12-2016

Sustainable management of lithium-ion batteries after use in electric vehicles

Kirti Richa
kxr7829@rit.edu

Sustainable management of lithium-ion batteries after use in electric vehicles

By

Kirti Richa

A DISSERTATION

Submitted in partial fulfillment of the requirements
for the degree of Doctor of Philosophy in
Sustainability

Department of Sustainability
Golisano Institute for Sustainability
Rochester Institute of Technology

December 2016

Author: _____
Sustainability Program

Certified by: _____
Dr. Callie W. Babbitt
Associate Professor of Sustainability Program

Approved by: _____
Dr. Thomas Trabold
Department Head of Sustainability Program

Certified by: _____
Dr. Nabil Nasr
Assistant Provost and Director, Golisano Institute for Sustainability and CIMS

Sustainable management of lithium-ion batteries after use in electric vehicles

By

Kirti Richa

Submitted by Kirti Richa in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Sustainability and accepted on behalf of the Rochester Institute of Technology by the dissertation committee.

We, the undersigned members of the dissertation committee, certify that we have advised and/or supervised the candidate on the work described in this dissertation. We further certify that we have reviewed the dissertation manuscript and approve it as partial fulfillment of the requirements of the degree of Doctor of Philosophy in Sustainability.

Approved by:

Dr. Callie W. Babbitt _____
(Committee Chair and Dissertation Adviser) Date

Dr. Gabrielle G. Gaustad _____
(Committee Member) Date

Dr. Nenad Nenadic _____
(Committee Member) Date

Dr. Brian Landi _____
(External Examiner) Date

SUSTAINABILITY PROGRAM
ROCHESTER INSTITUTE OF TECHNOLOGY
December 2016

ABSTRACT

Golisano Institute for Sustainability
Rochester Institute of Technology

Degree: Doctor of Philosophy **Program:** Sustainability

Name of Candidate: Kirti Richa

Title: Sustainable management of lithium-ion batteries after use in electric vehicles

In recent years, many forecasts have predicted a large scale adoption of electric vehicles (EVs), which would predominantly be powered by lithium-ion batteries (LIBs), owing to their high energy and power density and long cycle life. While use of EVs could reduce dependence on fossil based transportation fuels, there is a need to understand the end-of-life (EOL) implications of retired EV LIBs entering the waste stream in future in the battery-driven vehicle regime. To proactively address impending waste management issues and inform related policy, this dissertation explored the sustainable management of LIBs after use in EVs and the challenges and opportunities involved.

First, a future oriented, dynamic Material Flow Analysis (MFA) was conducted to estimate the volume of LIB wastes to be potentially generated in the US in near and long term. The objective of tracking future outflows of EOL EV LIBs through the MFA model was to: (a) Provide an understanding of the scale at which EV LIB waste management infrastructure needs to be developed in future, and (b) Analyze the composition of future EV LIB waste stream in terms of constituent LIB packs, cells and materials. The effect of EV adoption scenarios, variability in LIB lifespan distribution, battery energy storage, LIB chemistry and form factor on the volume, recyclability and material value of the forecasted waste stream was analyzed. Because of the potential "lifespan mismatch" between battery packs and EVs, LIBs with high reuse potential are expected in the waste stream. Results of the MFA model projected annual EV LIB waste flows of as high as 340,000 metric tons by 2040. Apart from the high volume, the projected EV LIB waste streams were characterized by the presence of a variety of recyclable metals, high percentage of non-recyclable materials, high variability in the potential economic value, and potential for battery reuse. Hence, a robust end of life battery management system would include an increase in reuse avenues, expanded recycling capacity, and safe disposal

routes accompanied by policy incentives to promote environmentally and economically favorable EOL management of EV LIBs.

Second, the environmental trade-offs of cascaded use of retired EV LIBs in stationary energy storage was investigated using cradle-to-grave life cycle assessment (LCA). The LCA model was framed from the dual perspective of stakeholders in the: (a) the EV sector, to understand if there is there a meaningful reduction in EV lithium ion battery environmental impact due to cascaded reuse, and (b) the Energy Utility sector, to understand if the utility sector could environmentally benefit from using refurbished EV lithium ion batteries for energy storage. In both the cases, an environmental benefit was obtained owing to avoiding the production and use of an incumbent lead-acid battery based system. However, there were diminished to no environmental benefits in scenarios where very few of the initial battery cells and modules could be reused and where service life was low in secondary application for refurbished EV LIB cells. Hence, environmental feasibility of cascaded use systems was found to be directly related to technical feasibility and reliability. An important methodological challenge addressed was the allocation of environmental impact associated with production and EOL management of LIBs across the EV and stationary use systems. The allocation modeling choices explored here were based on the concept of closed-loop recycling for material cascades. These modeling approaches can guide LCA of similar product cascade systems where a product is used for a cascaded second use in a different application.

Finally, a circular economy-inspired waste management hierarchy was proposed for EOL EVs from LIBs that included limited reuse in EVs, cascaded use in stationary applications, recycling and finally, landfill. To validate this circular economy approach, an eco-efficiency analysis was conducted across proposed waste management strategies for an EV LIB waste stream (modeled as 1,000 battery packs coming out of use in EV applications in the U.S.). Results demonstrated that a circular economy-centric waste management hierarchy can be environmentally and economically effective in managing the EV LIB waste stream in future, owing to benefits from reuse, cascaded use and recycling. However, such benefits would rely significantly on LIB size, testing procedures, the incumbent battery systems that used LIBs would displace, future prices of these batteries, and future recycling costs. Hence, these EOL management strategies would need policy and technology push to be viable. Although much attention has been placed on landfill disposal bans for batteries, results actually indicated that

direct and cascaded reuse, followed by recycling can together negate the eco-toxicity burden of unavoidable metal flows into landfill. When combined with regulations deterring landfill and policies promoting life cycle approaches that additionally consider design-for-EOL, battery maintenance, collection and safe transport, circular waste management systems can be improved for these batteries. Overall, a circular waste management system for EV LIBs is likely to complement existing and guide future policies governing EV LIB waste.

ACKNOWLEDGEMENTS

First and foremost, I would like to express my very special gratitude to Dr. Callie Babbitt, my Ph.D. advisor and mentor for her invaluable guidance and support throughout my doctoral journey. She has been an amazing role model for me during my graduate work and has been a continuous source of inspiration. Thank you for your constant encouragement, for challenging me and for pushing me forward throughout these years which has helped me grow as a researcher.

I would also like to thank my dissertation committee members Dr. Gabrielle Gaustad and Dr. Nenad Nenadic for their valuable insights during my dissertation work. The quality of my research papers greatly improved owing to the constructive feedback provided by Dr. Gaustad and the technical know-how of Dr. Nenadic.

I would additionally like to thank the Golisano Institute for Sustainability (GIS) for funding me during the first year of my Ph.D. and the National Science Foundation for subsequent years. A special thanks goes to the entire GIS family, faculty and staff, specially Dr. Nabil Nasr, Paul Stiebitz, Dr. Thomas Trabold, Dr. Eric Williams, Lisa Dammeyer, and Donna Podeszek.

I would further want to extend my gratitude to friends in India, US and Europe for making me believe that I can do it. Special thanks goes to my amazing friend Mugdha Patwardhan for always being there for me, during the difficult times and the happy days, for the constant support, the unending laughs and also, the car rides! I would also like to thank Mitesh Shah, for being a positive influence in my life which kept me going. Thanks Mitesh and Mugdha for being my family away from home and for being an immense source of motivation. Thanks Parin Maru for being such a sweet friend and for making the long trip to Rochester to attend my academic convocation.

Last, but not the least, I want to thank my family back in India for their unconditional love and constant faith in me. Mom, Dad and Ritwik, even though you all were thousands of miles away, I always felt your infinite support and the impact of your constant prayers.

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

CHAPTER 1: INTRODUCTION

1.1 Lithium-ion batteries

Lithium ion batteries (LIBs) were developed by Asahi Kasei Co. in Japan and were eventually commercialized by Sony Co. in 1991 (Yoshio et al., 2009). Owing to their high energy and power density and longer cycle and calendar life than the incumbent Nickel Cadmium (NiCd) and Nickel Metal Hydride (NiMH) battery technologies, LIBs quickly became very popular for use in consumer electronics such as cell phone and laptops.

In a typical LIB cell, lithium ions move between the anode and cathode, causing electric current. During charging, lithium ions are released from the cathode and move through a polymer separator to the anode. This enables the cell to store energy. During cell discharge, lithium ions in the anode move back into atomic-sized holes in the cathode material, thus producing energy. In both cases electrons flow in the opposite direction in an outer circuit. Owing to the reversibility of this chemical reaction, these cells are rechargeable (Linden & Reddy, 2003).

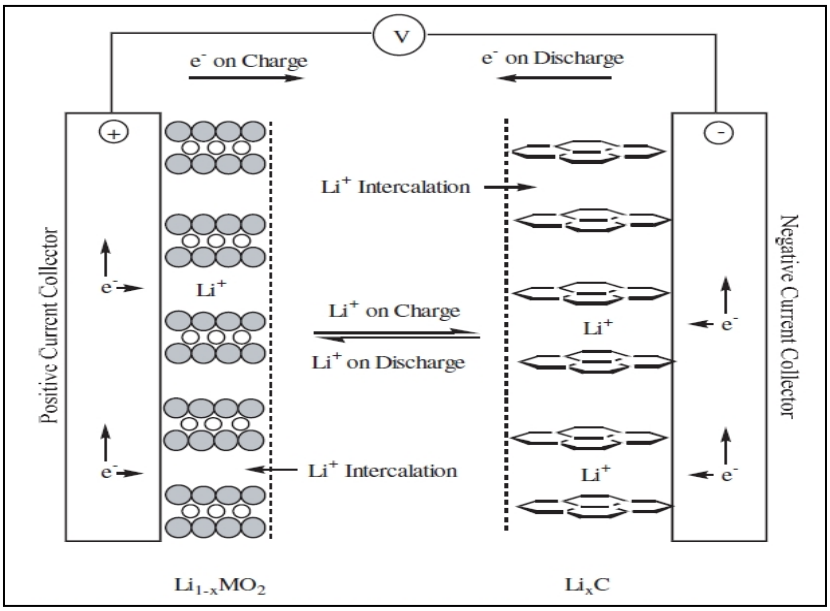


Figure 1.1 Schematic of the electrochemical process in a Li-ion cell (Linden & Reddy, 2003)

A lithium-ion battery cell contains four major components, viz. cathode, anode, electrolyte and separator. In general, the lithium-ion system consists of an inorganic lithium-intercalating compound as positive electrode, a lithium salt in an organic liquid as electrolyte,

and a lithium-intercalating negative electrode (generally carbon) (Vimmerstedt *et. al*., 1995). Table 1.1 lists some common materials used in lithium ion batteries.

Lithium-ion battery component	**Materials**
Anode	Carbon Graphite, Lithium Titanate ($Li_4Ti_5O_{12}$)
Cathode	Lithium cobalt oxide ($LiCoO_2$) Lithium nickel oxide ($LiNiO_2$) Lithium nickel cobalt oxide ($LiNi_{0.8}Co_{0.2}O_2$) Lithium nickel cobalt aluminum oxide ($Li_xNi_{0.8}Co_{0.15}Al_{0.05}O_2$) Lithium manganese nickel oxide ($LiMn_{0.5}Ni_{0.5}O_2$) Lithium manganese nickel cobalt oxide ($LiMn_{1/3}Ni_{1/3}Co_{1/3}O_2$; $LiMn_{0.4}Ni_{0.4}Co_{0.2}O_2$) Lithium manganese oxide ($LiMn_2O_4$) Lithium aluminum manganese oxide ($LiAlMnO_2$) Lithium iron phosphate ($LiFePO_4$)
Electrolyte solvent	Propylene carbonate (PC) Ethylene carbonate (EC) Diethyl carbonate (DEC) Dimethyl carbonate (DMC) Dimethoxyethane (DME) Ethyl methylcarbonate (EMC)
Electrolyte salt	Lithium hexafluorophosphate: $LiPF_6$ Lithium tetrafluoroborate: $LiBF_4$ Lithium(bis)trifluoromethanesulfonimide: $LiN(SO_2CF_3)_2$ Lithium tris (trifluoromethanesulfonyl)methide: $LiC(SO_2CF_3)_3$ Lithium trifluoromethanesulfonate: $LiCF_3SO_3$
Current collector	Aluminum, Copper
Separator	Polypropylene, Polyethylene

Table 1.1 Major components of lithium ion batteries and their common chemical constituents. Sources: Vimmerstedt *et. al*. (1995); Gaines and Cuenca (2000); Linden & Reddy (2003); Yoshio and Noguchi (2009)

1.2 Lithium-ion battery use in electric vehicles

The demand for consumer electronics is currently driving the LIB market with a global revenue-based market share of 60% in year 2013 (Frost and Sullivan, 2014). However over the last decade, these batteries have been introduced in electric vehicle (EV) applications with the Tesla Roadster being the first commercial battery electric vehicle (BEV) to be powered by LIBs (Berdichevsky et al., 2006). Owing to more than two decades of progress made towards improving the LIB technology both in terms of energy and power density as well as safety improvements (Howard & Spotnitz, 2007), they have become the preferred battery system adopted by leading EV manufactures like Chevrolet, Honda, Nissan, Ford, etc. While many hybrid electric vehicles (HEVs) in markets still use NiMH batteries, for plug-in hybrid electric vehicles and BEVs, LIBs are more attractive due to light weight, more than double the energy

density of NiMH batteries and four times that of lead acid batteries, longer cycle life, and the ability to provide deeper discharges.

Several agencies have predicted widespread diffusion of electric-drive vehicles in the future, both in the U.S. and at a global level. Forecasts of future EV sales (Figure 1.2) have been produced by the U.S. Energy Information Administration (EIA, 2012), J.D. Power and Associates (Humphrey *et al.*, 2010), Credit Suisse (Jobin *et al.*, 2009), International Energy Agency [IEA] (2011), Deutsche Bank (Watabe & Mori, 2011), Deloitte Consulting (Giffi *et al.*, 2010), Lazard Capital Markets (Shrestha *et al.*, 2010) and Morgan Stanley (Steinmetz & Shankar, 2008). The range of deployment scenarios by these agencies vary significantly across parameters (economic growth, oil price, proposed Corporate Average Fuel Economy [CAFE] standards, battery technology etc.), and indicate anywhere between 0.45 million to 4 million EVs sold in the United States in 2020 (Figure 1.2(a)) and international sales ranging between 5.2 million to 19.8 million in the same time frame (Figure 1.2 (b)). Powering these vehicles will clearly require a large scale deployment of lithium ion batteries (Gaines & Nelson, 2010; Gruber *et al.*, 2011; Kushnir & Sandén, 2012). In fact, it is predicted that in year 2020 EV batteries can account for about 30% of the LIB market shares (Frost and Sullivan, 2014), while the share of consumer electronics LIBs would drop from 60% to 24% (Figure 1.3).

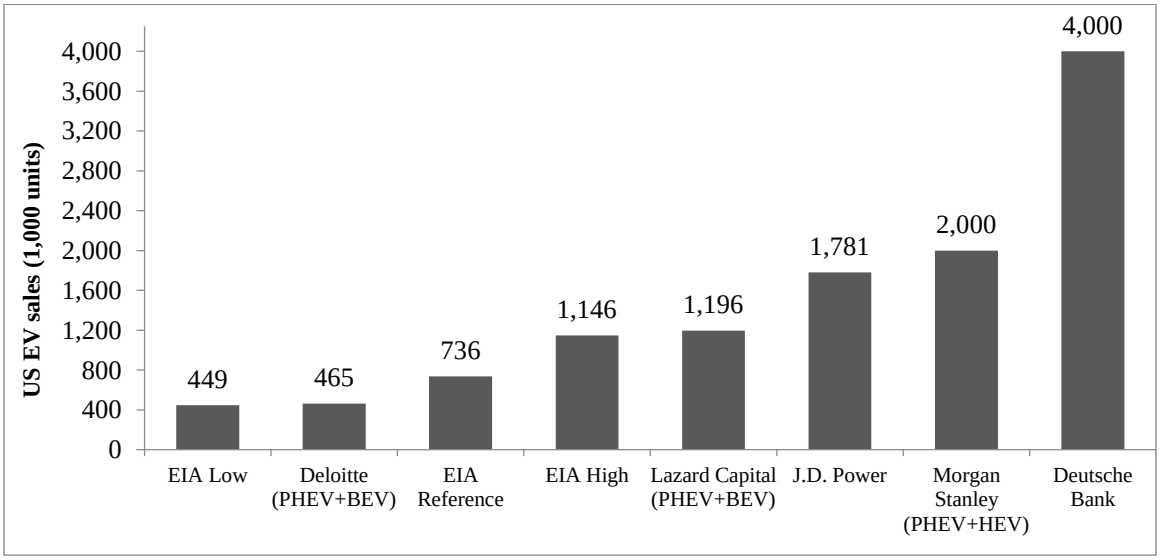


Figure 1.2 (a) EV sales forecast-2020 (U.S.). Literature references for each sales forecast provided in the main text.

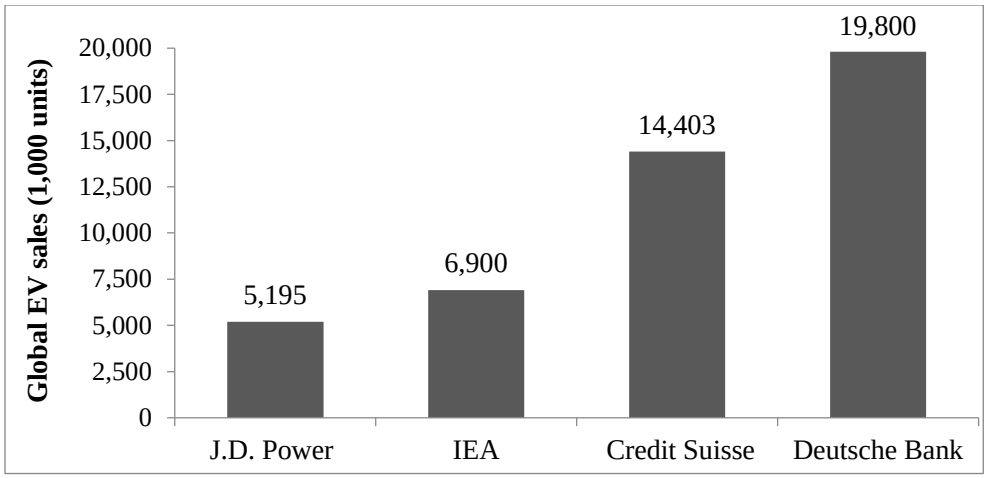


Figure 1.2 (b) EV sales forecast-2020 (Global). Literature references for each sales forecast provided in the main text.

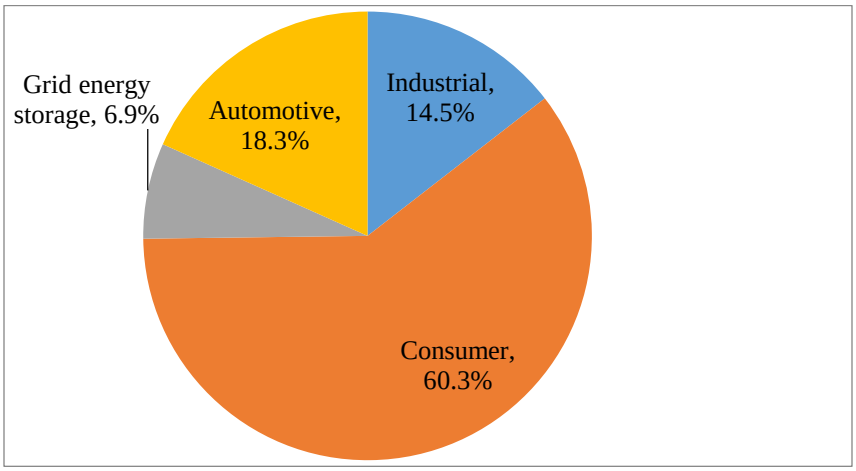


Figure 1.3 (a) LIB market revenue share by application in 2013 (Frost and Sullivan, 2014)

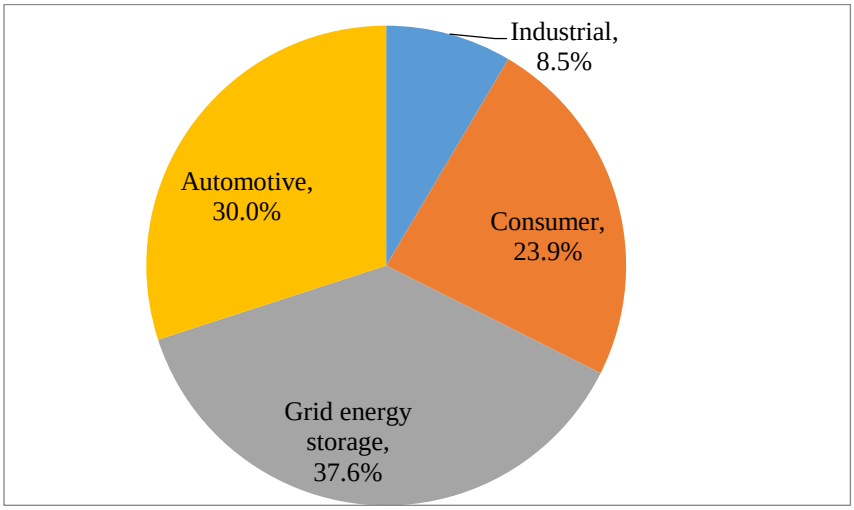


Figure 1.3 (b) LIB market revenue share by application in 2020 (Frost and Sullivan, 2014)

1.3 Sustainability implications

The rapid growth in LIB demand for EV application is likely to be accompanied with its own sustainability tradeoffs: as replacements for NiMH batteries, LIBs reduce demand for rare earth metals but increase consumption of lithium, cobalt, manganese and nickel (Alonso *et al.*, 2012; Gruber *et al.*, 2011). Several studies have investigated the implications of EV penetration on material demand, particularly lithium (Gaines & Nelson, 2010; Gruber *et al.*, 2011; Grosjean *et al.*, 2012; Kushnir & Sandén, 2012; Yaksic & Tilton, 2009). Though concerns over lithium scarcity in the long-term have been lessened by reassuring results from such studies, there may still be future challenges for the U.S. to access world lithium resources. A large portion of lithium deposits are found in only a few countries of the world, with the U.S. accounting for only 0.3% of current lithium reserves (USGS, 2012) and about 3.7% of the world lithium reserve base (USGS, 2009). Trade embargoes or political instability in the future may drastically impact the U.S. EV and LIB industries, as many lithium-supplying countries are already politically volatile. Furthermore, cobalt, manganese and nickel, which are major inputs to the lithium ion battery industry are not significantly mined in the U.S., leading to primary dependence on imported supplies (USGS, 2012).

Another major sustainability concern in the future would be the possibility of unchecked disposal of EV LIB waste that can cause environmental and health hazards, as demonstrated by unregulated electronic waste in past (Widmer et al., 2005; Robinson, 2009). The toxicity hazards caused by these batteries would depend on the materials comprising these batteries (Wang et al., 2014a). Due to the presence of metals like lithium, copper, cobalt, manganese, nickel, etc. the battery waste can create risk for soil and water contamination when disposed in landfills (Kang et al., 2013; Vimmerstedt, 1995). Currently the US EPA does not consider these batteries to be hazardous for the environment based on Toxicity Characteristic Leaching Procedure (TCLP) tests (Pistoia et al., 2001). A typical laptop battery consists of 6 to 9 LIB cells, but EVs would comprise of several hundred to thousands of cells per battery pack depending on the vehicle range (Berdichevsky et al., 2006; Nelson et al., 2011). The increase in magnitude of battery size as well as scale of battery production is expected to increase the extent as well as the probability of the hazards associated with these batteries. Hence, there is a need to analyze the health, environmental and safety hazards associated with them.

Many studies have investigated the life cycle environmental impacts of EV LIBs (Majeau-Bettez et al., 2011; Notter et al., 2010; Zackrisson et al., 2010; Samaras and Meisterling, 2008; Ishihara et al., 2002; Hawkins et al., 2013; Matheys et al., 2009, Ellingsen et al., 2013). LIBs have a much lower cumulative energy demand (CED) of virgin material in terms of MJ/Wh when compared to other battery chemistries such as nickel cadmium and nickel metal hydride batteries (Rydh and Sanden, 2005). However, some of the LIB materials still have relatively high CED and hence recovery of these materials would be beneficial from an environmental perspective (Table 1.2). Since lithium and manganese are currently not recycled at a commercial scale (Gaines, 2014; Wang et al., 2014b), the CED benefit of these metals in secondary form is yet to be established. EV LIB end-of-life (EOL) management strategies in terms of recycling and reuse would enable to deal with supply uncertainty issues of these materials and waste disposal concerns, as well as provide an environmental incentive by avoiding primary material mining and production.

LIB material	**CED –Primary metal (MJ/kg)**	**CED –Secondary metal (MJ/kg)**
Lithium	415	-
Aluminum	194	23.8
Nickel	187	14.7
Cobalt	128	38.4
Copper	60.5	28.1
Manganese	58.6	-
Steel	30.9	8.91

Table 1.2 Cumulative energy demand of some common LIB metals (ecoinvent Centre, 2010)

Moreover, recycling of EOL EV LIBs can be a valuable source of metal recovery. Forecasts estimate that EV LIB recycling markets can be worth more than 2 billion in year 2022 (Frost and Sullivan, 2010). However, the revenue from the EV battery recycling sector would depend on the cathode chemistry mix of this waste stream. Currently, most EOL batteries from consumer electronics contain high levels of cobalt, a metal whose high economic value catalyzes current LIB recycling systems, but the trajectory of battery technology could result in introduction of different material and value streams, which may change the economic and policy implications of battery recycling (Wang *et al.*, 2014a; Wang *et al.*, 2014b). For instance, lithium-iron phosphate and lithium manganese oxide batteries do not provide much economic incentive for recycling since recovery of battery grade manganese and lithium from these batteries proves to be more expensive than obtaining these metals from their ores (Gaines, 2014; Frost and

Sullivan, 2010; Wang et al, 2014b). However, economies of scale are expected in the future due to the large scale EV LIB waste to be generated that can drive recycling of these LIBs (Wang et al., 2014b). In case of lack of an economic incentive, policy mechanisms may be required to mandate or provide incentives to encourage recycling of low material value LIB waste streams.

Additionally, it is estimated that LIBs after the end of their useful life in EVs would have 70-80% of their capacity intact, thus capable of serving less demanding energy storage functions in the utility sector (Heymans et al., 2014, Williams and Lipman, 2010; Neubauer et al., 2012; Neubauer & Pesaran; 2011; Cready et al., 2003, Narula et al., 2011 etc.). Several economically and technically feasible secondary use possibilities for retired EV LIBs have been identified such as transmission support, light commercial load following, residential load following, and distributed node telecommunications backup power (Cready et al., 2003). Additionally, collaborations have been established between automobile manufacturers and utility providers to test the technical feasibility of EV LIB repurposing and "cascaded" use for stationary energy storage such as those between Nissan and Sumitomo Corporation or General Motors and ABB Group. In fact, a recent study by Sathre et al. (2015) demonstrated that second use of retired plug-in electric vehicles in California has the capability of delivering 5% of electricity demand of the state in year 2050. However, as in the case of recycling, the cascaded use model would be accompanied with its own obstacles in terms of the performance, reliability, technology and design requirements, business models, as well as lower perceived value by consumers (Neubauer & Pesaran, 2011; Cready et al., 2003; Frost and Sullivan, 2010; Hein et al., 2012). Moreover, since LIB cells have the potential safety threat of "thermal runaway", the cascaded use pathway can face additional regulatory barriers governing the shipping and collection of EV LIBs and siting of large stationary energy storage systems (Elkind, 2014). Overcoming these roadblocks and economic and technical constraints of EV LIB secondary use in stationary application can create a sustainable market of repurposed EV LIBs grid-based, off-grid and renewable energy storage applications.

Irrespective of the barriers and concerns associated with recycling and reuse pathways of EV LIBs, both the EOL management routes provide the possibility of reducing net environmental impacts of these batteries by reducing battery waste deposition as well as avoiding resource depletion, energy use and other environmental impacts (e.g. toxicity, greenhouse gas emissions etc.) associated with production of LIBs or their constituent materials.

From a systems perspective, both cascaded use of whole LIB packs, module or cells and recycling of constituent LIB materials have the potential of magnifying the environmental gains from use of EV technology (Ahmadi et al., 2014a). Since LIBs constitute a major cost component of EV ownership, development of EV LIB reuse avenues and economically feasible recycling technology for closing the loop of LIB materials has the ability of reducing the battery cost and encouraging EV adoption.

1.4 Dissertation outline

This dissertation aims to analyze the environmental and economic implications of EOL EV lithium-ion batteries by:

(1) Estimating and characterizing the EV LIB outflows potentially entering the waste stream due to their increasing deployment in electric vehicles over the short and long term future in the United States (Chapter 2)

(2) Analyzing the life cycle environmental benefits of cascading batteries from the EV waste stream into a second use in stationary energy storage applications (Chapter 3)

(3) Assessing the environmental and economic trade-offs of different end-of-life management pathways of EV LIBs along a circular economy inspired waste management hierarchy (Chapter 4)

To achieve these objectives, a combination of techniques from industrial ecology were applied, including material flow analysis and life cycle assessment, coupled with scenario analysis, systems analysis, cost-benefit analysis and empirical approaches.

As a proactive step towards understanding future waste management challenges, Chapter 2 presents a future oriented material flow analysis (MFA) used to estimate the volume of LIB wastes to be potentially generated in the United States due to EV deployment in the near and long term future. Because such an analysis is complicated by significant uncertainty about technology adoption and performance, this MFA is also informed by approaches used in previous studies to develop scenario-based MFA for materials ranging from steel (Park *et al.*, 2011; Pauliuk, Wang, & Müller, 2011; Michaelis & Jackson, 2000) to electronic waste (Steubing *et al.*, 2010; Kang & Schoenung, 2006; Streicher-Porte *et al.*, 2005). From a methodological standpoint, Chapter 2, thus, also seeks to highlight the uncertainties associated with conducting a scenario-based MFA of EV LIBs, as a means of establishing future research priorities that must

be resolved as additional data and system parameters become available. Key uncertainties addressed here include EV adoption dynamics, battery lifespan and constituent LIB cells. Hence, a set of scenarios was developed to bound the parameters most influential to the MFA model and to forecast "low," "baseline," and "high" projections of future EOL battery outflows from years 2015 to 2040. These models were implemented using technology forecasts, technical literature, and bench-scale and battery modeling data characterizing battery material composition. The waste stream under the different modeling scenarios was analyzed for material volume, reuse potential, recyclability and material value. By highlighting the timing, variety and volume of materials expected in the future EV LIB waste stream, the MFA model would help policy makers to develop proactive measures for EOL battery reuse, recycling and safe waste disposal. Additionally the EV-LIB MFA system in Chapter 2 presents a unique case of MFA modeling of "dual-product systems" where a lifespan mismatch is expected between EV and the LIBs. This MFA therefore serves as a model to analyze waste flows and obsolescence dynamics of similar systems such as photovoltaic cells and their parent-modules, electronic equipment and parts or automobiles and their components.

To build on the several techno-economic analyses of EV LIB cascaded second use in stationary energy storage (Neubauer and Pesaran, 2011; Neubauer et al., 2012; Cready et al., 2003; Williams and Lipman, 2010; Viswanathan et al., 2011; Narula et al., 2011, etc.), in Chapter 3 the environmental tradeoffs from cascaded use of retired EV LIBs in stationary energy storage application was analyzed. A cradle-to-grave life cycle assessment (LCA) approach was employed to jointly model two systems to address the consideration of stakeholder groups corresponding to both first (EV) and second life (stationary energy storage) battery applications. The environmental feasibility criterion was defined by an equivalent-functionality lead-acid (PbA) battery which is the incumbent technology widely adopted for stationary energy storage applications (Soloveichik, 2011). The dual-stakeholder approach was adopted to understand the potential for dual benefit from EV LIB second use– both from the perspective of offsetting initial manufacturing impacts by extending the lifespan as well as avoiding production and use of a PbA battery system. The LCA model also tested sensitivity to parameters such as the fraction of battery cells viable for reuse, service life of refurbished cells, and PbA battery efficiency. A critical methodological challenge addressed in Chapter 3 was the allocation of environmental impacts associated with producing LIBs across the EV and stationary use systems.

In Chapter 4, a waste management hierarchy inspired by circular economy principles was proposed for EOL management of retired EV LIBs entering the US waste stream in year 2030. Four EOL management pathways were modeled: 1) a small fraction of LIBs would meet technical requirements for limited **reuse** in used EVs, 2) a majority of used LIBs would be directed to "**cascaded use**" in a secondary grid energy storage application, 3) non-reusable batteries would be **recycled**, and 4) all remaining materials would be **landfilled**. In fact, the EU Waste Framework Directive (2008/98/EC) promotes the circular economy concept and recommends a waste management hierarchy of prevention, reuse, recycling, energy recovery and disposal, with prevention and reuse the preferred waste management approaches (European Parliament, 2008).

The environmental and economic implications of different EOL pathways of waste electronics have been explored in the past (Wang and Gaustad, 2012; Iakovou et al., 2009; Williams et al., 2008; Kiritsis et al., 2003; etc.) to guide policies to include both regulatory interventions and economic incentives for producers to take up extended producer responsibility. In general, a waste management hierarchy depicts priorities from an environmental, as well as from an economic perspective for electronics (Brandstotter et al., 2004) but it is yet to be analyzed for LIBs. While the European Union and few states in the US ban the landfill of batteries (CA Code, 2006; New York State Rechargeable Battery Law, 2010, European Commission, 2006), waste regulations in both EU and US can potentially be expanded to more specifically address management of LIBs from vehicles along different EOL pathways. Since a comprehensive eco-efficiency analysis of EV LIB waste management routes is currently absent, a lack of understanding of the economic costs or benefits of the different EOL management pathways of LIBs may delay proactive policy instruments to be implemented to facilitate the adoption of the environmentally preferable battery waste management route. Hence, in Chapter 4, a case study was developed to examine the eco-efficiency of the proposed waste management framework along metrics like cost savings, cumulative energy demand (CED), eco-toxicity, and metal depletion. Since EOL management pathways for LIBs along the proposed hierarchy must also consider policy implications, gaps in current policies were identified and the results of the case study were used to set a roadmap for EV battery EOL management research and policy to improve the "circularity" of the proposed system.

CHAPTER 2: A FUTURE PERSPECTIVE ON LITHIUM-ION BATTERY WASTE FLOWS FROM ELECTRIC VEHICLES

2.1 Introduction

Lithium-ion batteries (LIBs) have emerged as a promising energy storage solution for electric vehicles (EVs) and renewable energy systems, but their potential environmental tradeoffs are not well characterized. Although recent work has focused on supply side issues, such as lithium availability, key uncertainties surround the emergence and management of these batteries in the waste stream and the ability of domestic recycling infrastructure to recover scarce and valuable materials from a highly variable mix of discarded batteries. A proactive approach is required to prevent unanticipated environmental impacts of end-of-life (EOL) battery generation associated with forecast growth in electric vehicle deployment.

Clearly, a better understanding of the ultimate management and fate of batteries in the waste stream is required, but such an analysis is complicated by key uncertainties, including the expected timing and volumes of batteries reaching their end of life; the quality, concentrations, and variability of specific materials contained in spent batteries; and the capacity for recycling systems to recover scarce and valuable materials from a highly variable battery waste stream. While the lag in deploying EV technologies may suggest that battery waste will not be a priority for several years, "lessons learned" from our current sub-optimal management of electronic waste show the perils of introducing complex products without proactive development of a waste management system. In the case of electronic waste, low end-of-life value, difficulty recovering valuable materials and insufficient domestic infrastructure has lead to exploitation of developing countries and loss of valuable material resources (Babbitt *et al.*, 2011; Williams *et al.*, 2008; Widmer *et al.*, 2005; Wang & Gaustad, 2012). Since many of these factors are similar to LIBs, avoiding negative environmental, economic, and social outcomes at EOL requires a more proactive approach in planning for this new waste stream.

As a step towards addressing EOL LIB management, this chapter applies a scenario-driven material flow analysis (MFA) to project the potential volume and timing of lithium-ion batteries entering the waste stream as a result of their forecasted deployment in electric vehicles. Towards this objective, the number of EV LIB units entering the waste stream as well as the mass of battery cells in that stream is estimated on an annual basis between years 2015 and 2040 for three different scenarios. To estimate recycling potential and waste management needs of

EOL EV LIBs in the future, this chapter also aims to characterize the materials that would be present in the EV battery waste stream on the basis of their recyclability and their commodity value under different technology trajectories of battery chemistry and form factor. Furthermore, this MFA model also seeks to characterize the potential for diverting EV batteries from this waste stream into reuse applications depending on the remaining battery life.

MFA is a well-established method for investigating the material, energy and environmental implications of commodity products (Oguchi *et al.*, 2008; Yoshida *et al.*, 2009; Chang *et al.*, 2009; Steubing *et al.*, 2010 etc.). While some MFA studies have addressed issues related to LIBs, the existing literature focuses on analyzing the stock and flows of laptops and cell phones batteries (Chang *et al.*, 2009), tracking flows of cobalt (Harper *et al.*, 2012), and assessing supply and demand for lithium due to EV technology (e.g., Gaines and Nelson, 2010; Grosjean *et al.*, 2012; Kushnir and Sandén, 2012). As such, no study has yet applied MFA to fully model future outflows of batteries from EV systems.

2.2 Method

A future oriented top-down MFA was conducted to estimate the volume of lithium-ion batteries projected to enter the waste stream in the near and long term future, after use in electric vehicles. MFA is a systematic assessment of the flows and stocks of materials within a defined temporal and spatial system (Brunner & Rechberger, 2004) that can be used to track the flow of a specific substance or of products within a system. In the top-down MFA methodology, the product inflows are determined from specific 'final goods' categories entering the system and the outflows are determined from discards, based on product lifespan, with the material stocks being inferred from these inflows and outflows (Graedel & Allenby, 2010). Here, the annual inflow of EV batteries was estimated from projected EV sales, and the annual outflow of batteries was calculated based on battery and vehicle lifespans. Given the significant uncertainty about future EV adoption rates and battery technologies, bounding scenarios were developed to forecast "low," "baseline," and "high" projections of future waste battery outflows and their attendant material implications. Key differences among these scenarios stemmed from variability in EV sales projections, battery lifespan distribution and parameters governing number of cells per battery pack, which will be discussed in the subsequent sections.

2.2.1 Model formulation

The EV battery MFA model was implemented in three sequential steps, each described in more detail in the following sections:

(1) Material flow analysis to estimate the waste flows of entire EV battery packs

(2) Estimation of individual lithium-ion cells contained in the EV battery pack waste stream

(3) Assessment of specific materials comprising each cell within the EV battery waste stream

Furthermore, based on the material and mass composition of the EV battery waste stream, the economic value of the waste stream was estimated on an annual basis.

Material flow analysis to estimate the flows of waste EV battery packs: The first part of the model calculated the number of lithium-ion EV battery packs entering the U.S. waste stream on an annual basis from years 2015 to 2040. This time period was chosen based on available data from the Department of Energy on both near- and long-term EV deployment projections. The annual inflow of EV batteries was estimated from EV sales forecasts, and the annual outflow of waste batteries was determined based on the battery lifespans once they entered vehicle use, as well as the lifespan of the EV itself.

EV sales forecast: U.S. level EV sales forecasts were obtained from the Light Duty Vehicle (LDV) Sales Projections through the year 2035 provided by the U.S. Energy Information Administration [EIA] (EIA, 2012). Three types of electric-drive vehicles were considered in the EIA LDV sales forecasts: hybrid- [HEVs], plug-in hybrid- [PHEVs] (10 miles and 40 miles ranges) and all-electric or battery electric vehicles [BEVs]. For the baseline scenario, the EIA Annual Energy Outlook “reference case” LDV sales projections were used. The reference case used in EIA projections is a baseline scenario assuming business-as-usual with current laws and regulations being the same across the timeline of the projections (EIA, 2012). The low and high scenarios reflect EIA forecasts that consider low and high oil prices, respectively. These forecasts are shown in Figure 2.1.

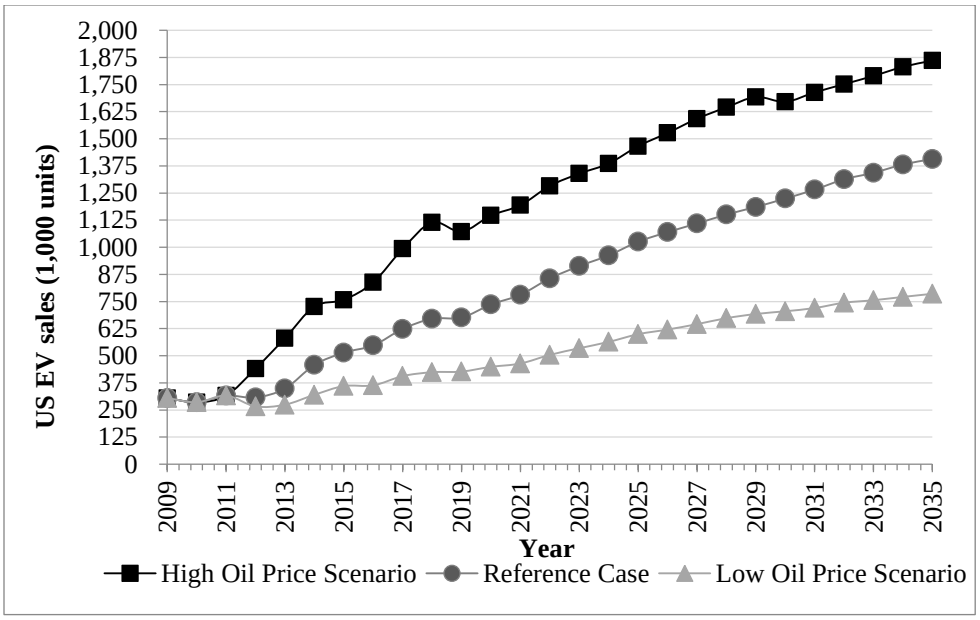


Figure 2.1 Energy Information Administration (2012) EV sales forecasts

Battery lifespan: The lifespan or service life of a lithium-ion battery can be expressed either in terms of its cycle life or its calendar life. Cycle life is defined as the number of charge-discharge cycles the battery can undergo before failing to meet specific performance criteria. Calendar life on the other hand is defined as the length of time a battery can be stored with minimal discharges before capacity diminishes. In general, a battery is considered to have reached its end of life in EV application when it reaches about 80% of its original capacity (William and Lipman, 2010). EV battery lifespan is highly uncertain and dependent on many factors which are still poorly understood. Marano *et al.* (2009) indicates that lithium-ion batteries usually have a calendar life of 10 years, subject to favorable operating conditions that avoid overcharging, aggressive driving leading to rapid discharge and more frequent charging, and operation at high temperatures. Most previous studies have assumed a fixed EV battery lifespan of either 8 or10 years (Gruber *et al.*, 2011; Yaksic & Tilton, 2009; Harper *et al.*, 2011), which is consistent with the length of many vehicle manufacturers' warranty terms. However, some literature indicates lower lifespan of about 5 years for EV LIBs (Anderman, 2007). As per Dinger *et al.* (2010), EV battery life span could be anywhere between 5 to 10 years, while Nemry *et al.* (2009) assume a lifespan of 10-15 years. Significant research efforts are aimed toward achieving a higher lifespan for almost 15 years for EV batteries (Kalhammer *et al.*, 2009; Chalk & Miller, 2006).

Applying a lifespan distribution to determine the EV-LIB outflows would address the fact that the lifespan of a battery would depend on its usage and charging patterns, which vary from

user to user. Assuming that electric vehicles are charged 1.5 times per week, the U.S. Department of Energy (U.S. DOE, 2010) predicted that the calendar life of a typical EV battery would increase from 4 years in 2009 to 14 years in 2015 owing to the ongoing innovation in this field. Hence, the different scenarios in the model have considered battery life spans ranging from 4 to14 years. Rather than a single point estimate, a lifespan distribution (Figure 2.2) was applied to model a more realistic scenario, taking into consideration early battery failures as well as batteries surviving for more than 10 years. Since a lifespan distribution of EV LIBs is not yet established, this technology being in its early stages of adoption, a truncated normal distribution of EV LIB lifespan has been used in the three scenarios (with a mean lifespan of 8 to 10 years). The variation in assumed battery lifespan distribution among the three scenarios not only indicate the uncertainty in the lifespan of EV LIBs but also highlight that the volume of EV battery waste stream would be dependent on battery lifespan to a certain extent. For instance, in the low scenario, 70% of EV LIBs have been assumed to have lifespan exceeding 8 years, whereas this percentage is 50% and 35% respectively for batteries in the baseline and high scenarios respectively. In spite of these variations, the distributions selected result in a majority of EV LIBs used in EVs modeled as having a lifespan in the range of 8 to 10 years, consistent with warranty terms and recent literature.

Similar to EV batteries, the lifespan of electric vehicles too would follow a distribution which may be even wider than that for batteries, depending on early vehicle failure or car crashes as well as extended life through multiple resales. However, to keep this initial MFA model tractable, the EV lifespan has been fixed. In general, traditional vehicle lifespan assumptions vary across studies in the range of 10 to 16 years (Huang *et al.*, 2011; Greene & DeCicco, 2000; Lemp & Kockelman, 2008; Greene *et al.*, 2005; Kumar & Sutherland, 2008). Only limited information is available on electric vehicle lifespan, but this parameter is modeled as 10 years in a recent study by Gruber *et al.* (2011). While the uncertainty associated with lifespan and the need for future work in this area is recognized, this MFA model assumed a moderate, fixed EV lifespan of 10 years as a starting point for analysis, with sensitivity analysis on a 16 year EV lifespan shown in section S7.3 of Appendix A.

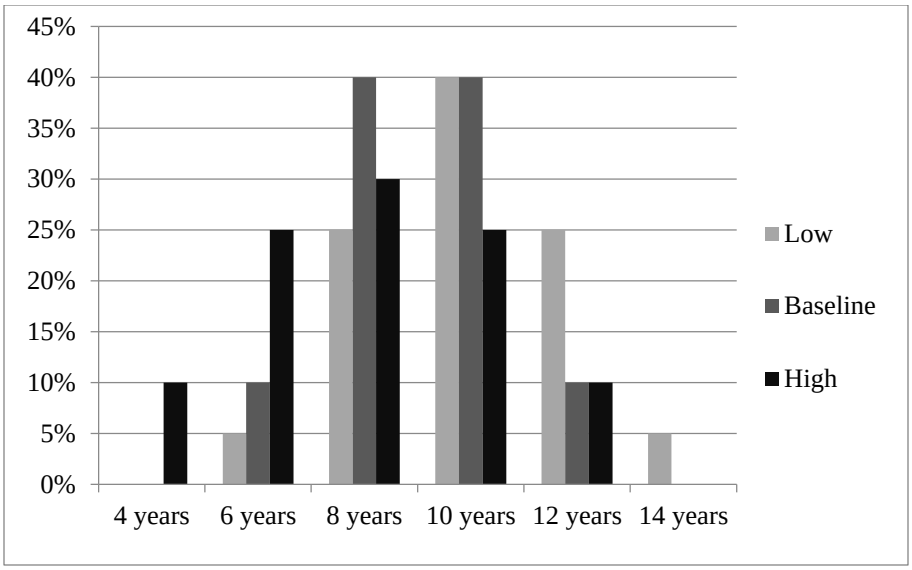


Figure 2.2 Truncated lifespan distribution of EV batteries for three scenarios

The lifespan distribution shown in Figure 2.2, contrasted against the vehicle life, raises an important point: there will likely be a "mismatch" between vehicle and battery lifespans. Some batteries entering use in a given year would likely reach the end of their life before the vehicles in which they are used. These vehicles then need new batteries to continue operation in subsequent years. On the other hand, if a vehicle were to reach the end of its life before its batteries, it is assumed that the battery would not be refitted into a new vehicle (although it may be reused in other applications) (Williams & Lipman, 2010; Cready *et al.*, 2003). Thus, batteries entering the waste stream at any given time can be loosely classified into two types:

Type 1 EOL EV batteries are those that have reached their end-of-lives in EV application due to capacity fade, either before or coinciding with the vehicles' end of life. In general, an EV battery has 70-80% of its original capacity intact once it reaches the end of its utility for EV applications (Neubauer & Pesaran, 2011). Though insufficient for automotive use, there is some potential that these batteries can be reused in off-grid and grid-based stationary energy storage applications instead of entering the waste stream (Neubauer & Pesaran, 2011; William & Lipman, 2010; Cready *et al.*, 2003).

Type 2 EOL EV batteries are those found in vehicles that reach their end-of-lives before their batteries, which is likely the case in early vehicle failure or crash or if a vehicle has a battery replacement later in its useful life. This set of non-EOL EV batteries could technically still meet the criteria for reuse in EVs, but actual reuse in this manner is unlikely, given concern about reliability and technical compatibility of "pre-aged" batteries (Cready *et al.*, 2003; Burke,

2009). These batteries may have high potential for other reuse markets, like those described above. The distinction between these two battery types is intended to indicate the potential for diverting batteries from the waste stream into reuse applications.

Lithium-ion battery use in hybrid electric vehicles

Currently most HEVs on the market use nickel metal hydride [NiMH] batteries, rather than lithium-ion, and NiMH batteries would continue to be a feasible option for HEV for several years (Frost & Sullivan, 2009). However, it is predicted that lithium-ion batteries' share of the HEV market would grow and eventually surpass NiMH usage between 2018 and 2025 (Jobin *et al.*, 2009; Madani, 2009; Fu, 2009). Estimates from a Credit Suisse report prepared by Jobin *et al.* (2009) were applied to the scenarios used here, as their study provided both conservative and optimistic estimates for HEV lithium-ion battery adoption. The high and baseline scenarios started from the Credit Suisse bottom-up estimates, which were optimistic towards rapid LIB adoption in HEV (Jobin *et al.*, 2009), leading to an assumption that all HEVs use lithium-ion batteries by year 2015 and 2025, respectively in these two scenarios. The Credit Suisse top-down estimate for HEV lithium-ion battery adoption was used for the low scenario, with an assumption that 100 percent of HEVs would not rely on lithium-ion batteries until the year 2032. Details about HEV lithium-ion battery adoption are provided in Appendix A.

Estimation of EV battery pack outflows

Considering the sales and lifespan assumptions stated above, Figure 2.3 illustrates the conceptual basis of estimating EV battery outflows by this model:

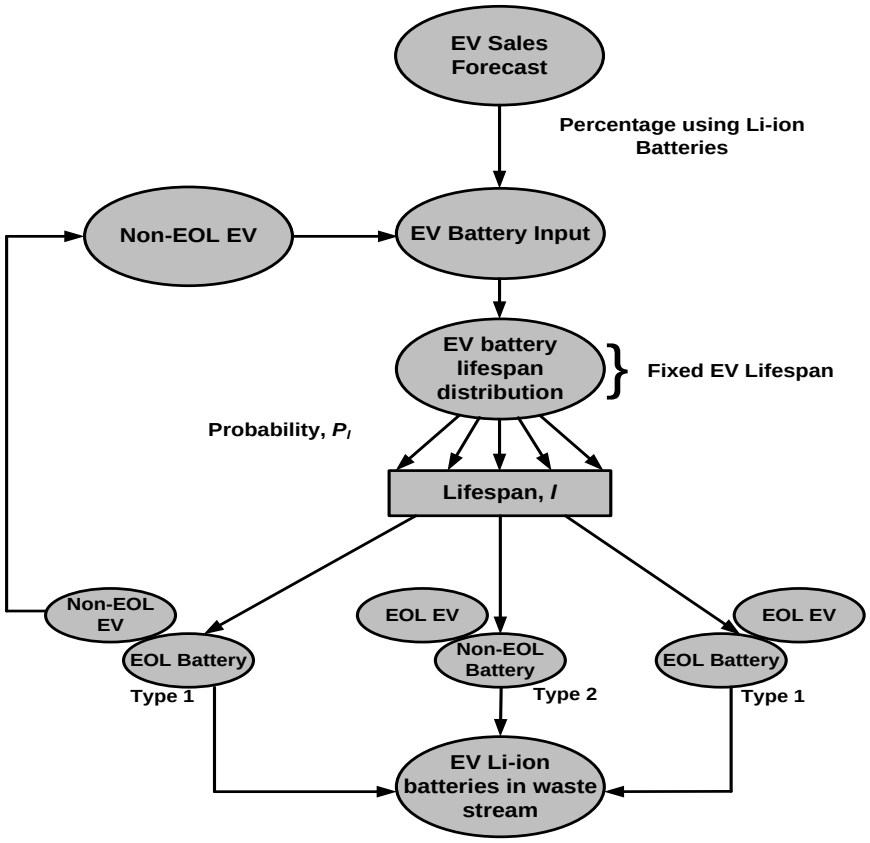


Figure 2.3 Conceptual basis of estimating future EV Li-Ion battery outflows

The number of new LIBs entering EV use in any year *t* would depend on EV sales in year *t*, as well as the number of non-EOL EVs which would require a replacement battery in that year (Figure 2.3). Here, it was assumed that all non-EOL EVs would use a replacement battery, while it is recognized that realistically, all vehicles may not be put back into use due to high replacement battery cost or damages due to automotive accidents. The lifespan distribution was based on "P_l", the percentage of batteries sold in any given year to have a useful life of *l* years in EV application, which varied based on the scenario (Figure 2.3). K_t, the total number of lithium-ion battery packs entering use in EVs in year *t* was determined as follows:

$$K_t = \sum_i (S_{i,t} + W_{i,t}) \quad (1)$$

$S_{i,t}$ = Sales of new EVs of type *i* that use LIBs in year *t*

$W_{i,t}$ = Non-EOL EVs of type *i* requiring a replacement LIB in year *t*

The above relationship was distinguished by "*i*" vehicle types: BEV, PHEV10, PHEV40, and HEV (the percentage of HEVs that use lithium-ion batteries). The number of batteries entering new EVs ($S_{i,t}$) was determined by the sales forecast for that year, as described in a

previous section. The non-EOL EVs requiring a replacement battery ($W_{i,t}$) was based on the scenario-specific cases of first-use batteries with a shorter lifespan than the vehicles in which they were used. In cases of extreme "lifespan mismatch," vehicles with very long lifespans paired with batteries with very short lifespans may require two battery replacements. Hence, for a given year, *t*,

$$W_{i,t} = \sum_{i}\sum_{l} P_l * (S_{i,(t-l)} + W_{i,(t-l)}) \qquad (2)$$

l= EV battery lifespan,

s.t. *l*< EV lifespan,

P_l = Percentage of EV LIBs sold in any given year to have a useful life of *l* years in EV application

$S_{i,(t-l)}$ = Sales of new EVs of type *i* in year (*t*-*l*)

$W_{i,(t-l)}$ = Non-EOL EVs of type *i* requiring a replacement battery in year (*t*-*l*)

Thus, the number of EV LIB packs entering the waste stream (*B*) in a given year *t* after an *l* year lifespan is expressed as:

$$B_t = \sum_{i}\sum_{l} P_l * (K_{i,(t-l)}) \qquad (3)$$

$K_{i,(t-l)}$ = Total number of LIB packs entering use in EVs of type *i* in year (*t*-*l*)

Estimation of individual lithium-ion cells contained in the EV battery pack waste stream

The approach described thus far focused on total battery packs, which each may contain a varied number and type of cells, depending on technical specifications such as EV type and cathode chemistry. Next, the number of lithium-ion cells in this EV battery waste stream was estimated, for a given year *t* as

$$N_t = B_t * \sum_{i}\sum_{j} (PE_{i,t} * PC_j * D_{i,j}) \qquad (4)$$

B_t =Number of LIB packs in EV battery waste stream in year *t*

i=EV type (BEV, PHEV10, PHEV40, HEV)

j= LIB cathode chemistry

$PE_{i,t}$= Percentage of waste LIB packs belonging to EV type *i* in year *t*

PC_j= Percentage of LIBs of battery chemistry *j* in EV battery waste stream

$D_{i,j}$=Number of cells per LIB pack, specific to EV type and cathode chemistry

Parameters B_t and $PE_{i,t}$ change with time as well as with the scenario under consideration as they are functions of annual EV sales within a given scenario. On the other hand, PC_j and $D_{i,j}$ were assumed constant with time, though $D_{i,j}$ does vary across the scenarios as shown in Table S3.2 in Appendix A.

The number of battery packs in a given year *t* (B_t) was obtained from the EV battery MFA results discussed in the previous section. $PE_{i,t}$, percentage of waste batteries belonging to a given EV type in year *t* was based on the relative prevalence of each type of EV sold, and thus entering the waste stream. Four prevalent lithium-ion cathode chemistries (i.e. *j*) were considered, namely, lithium cobalt oxide ($LiCoO_2$), lithium manganese oxide ($LiMn_2O_4$), lithium iron phosphate ($LiFePO_4$) and lithium nickel cobalt manganese (NCM) oxide, all having 18650 form factor cells (cylindrical cells with 18 mm diameter and 65 mm length). The selection of this form factor was based on data availability, with the recognition that results may change with alternative form factors, like the prismatic cells, expected to be used in most EVs. Sensitivity analysis was conducted on this assumption as described in following sections. While the current LIB waste stream is almost entirely made up of consumer electronic batteries, which typically contain 100% $LiCoO_2$ cathode chemistry (Wang *et al.*, 2014b), the distribution of cathode chemistries assumed in this chapter (i.e. PC_j) for all three scenarios is 10% $LiCoO_2$, 30% $LiMn_2O_4$, 30% $LiFePO_4$, and 30% NCM. This distribution was selected to reflect that all three latter cathode chemistries are likely candidates to replace the existing lithium cobalt oxide based batteries for EV application, aside from limited application as in the case of Tesla vehicles. The number of cells per battery pack for a given EV type using a given battery chemistry ($D_{i,j}$) varied with the scenario under consideration as described in the following section.

Determination of number of cells per battery pack ($D_{i,j}$)

The number of cells per LIB pack for a given EV type and a given battery chemistry ($D_{i,j}$) was estimated from the energy storage capacity of the EV battery pack ($Epack_i$), dependent on the EV type *i* and the energy storage of individual cells ($Ecell_j$), dependent on the cell cathode chemistry *j* and was estimated as follows:

$$D_{i,j} = (Epack_i(Wh) / Ecell_j(Wh)) \quad (5)$$

Battery energy storage ($Epack_i$)

The battery pack energy storage ($Epack_i$) depends on the EV type and its associated electric range, as well as other parameters like vehicle consumption rate and percent battery efficiency and available energy:

$$Epack_i = (R_i * C)/(\eta * A_i) \quad (6)$$

R_i =Electric range of EV type i (miles)

C = Electric vehicle consumption rate (Wh/miles)

η= Percent efficiency of EV LIB

A_i = Percent available energy of the total EV LIB energy for a given EV type i

While the vehicle electric range remains constant with each scenario, the other three parameters in Equation 6 will vary over the three scenarios:

(1) Vehicle electric range (R_i)

The electric ranges for the three EV types were determined based on EIA (2012) and Gaines and Nelson (2010) and were kept fixed across all scenarios. The BEVs were assumed to have 100 miles electric range, the HEVs were assumed to have 4 miles electric range. In case of PHEVs, both 10 and 40 miles electric ranges were considered.

(2) Vehicle consumption rate (C)

The consumption rate of an electric vehicle can be defined as the electrical energy consumed per mile of travel. Table S5.1 in Appendix A lists the energy consumption rates of electric vehicle models in the recent years (according to EPA tests), and assumptions documented in the literature. Based on these values, the consumption rate of EVs was assumed to be 250 Wh/mile, 300 Wh/mile and 350 Wh/mile for the low, baseline and high scenarios, respectively.

(3) Battery efficiency (η)

The overall energy stored by the battery available for electric vehicle application depends on the energy efficiency of the battery, so there is an inverse relationship between efficiency and number of cells. The battery efficiency determines the amount of energy taken out during

discharge after it was initially charged. The most common energy efficiency value for lithium-ion batteries reported in literature is 90% (Van den Bossche *et al.*, 2006; Gondelach, 2010; Shiau *et al.*, 2009; Karden *et al.*, 2007; Tanaka *et al.*, 2001; Matheys *et al.*, 2008). For the Tesla Roadster BEV, the efficiency of the charge-discharge cycle of lithium-ion batteries was reported to be approximately 86% (Eberhard & Tarpenning, 2006). Campanari *et al.* (2009) have assumed a 92% efficiency of lithium-ion batteries used in electric vehicles. According to Rydh and Sandén (2005), the efficiency of lithium-ion batteries can lie anywhere between 85% and 95%. Hence, the EV battery efficiency was assumed to be 95%, 90%, and 85%, respectively for the low, baseline, and high scenarios (see Table 2.1).

(4) Available energy of EV battery (A_i)

The available energy of an EV battery is typically less than the total energy stored because the depth of discharge is restricted to preserve battery life and for safety purposes (Axsen, Burke, & Kurani, 2008). According to Srinivasan (2008), the available energy of a HEV battery is 20-30% of its total energy, while for a BEV or PHEV battery it could be as high as 70-80%. As per the Argonne National Laboratory [ANL] (2012) BatPaC model, the energy utilized by a HEV battery is 25 % of the total energy, while it is 70-75% and 85-90% for a PHEV and BEV battery respectively. Based on these ranges, assumptions for the available energy percentage of the total battery energy for each of the vehicle type and for the three scenarios are as shown in Table 2.1, which also includes assumptions for the vehicle consumption rate, battery efficiency and available energy for the three scenarios, and the calculation of EV battery energy storage based on these factors:

Scenario	Vehicle Consumption Rate (Wh/mile)	Battery Efficiency	Battery Available Energy (Percentage)			EV Battery Energy Storage (kWh)			
			BEV	PHEV	HEV	BEV	PHEV10	PHEV40	HEV
Low	250	95%	90%	80%	30%	29.2	3.3	13.2	3.5
Baseline	300	90%	85%	75%	25%	39	4.4	18	5.3
High	350	85%	80%	70%	20%	51	5.9	24	8.2

Table 2.1 Vehicle consumption rate, battery efficiency, percent available energy and EV battery energy storage for the three scenarios

Cell energy storage ($Ecell_j$)

The energy storage of 18650 cells for the four battery chemistries considered was obtained as the product of cell capacity and the nominal or average cell voltage as described in section S3 in Appendix A. The cell capacity (mAh) was estimated as the product of the cathode mass and the specific capacity (mAh/g) of lithium-ion cells for each of the four cell chemistries considered in the model. The specific capacity of the lithium-ion cells was obtained from Dahn and Erlich (2011). The cathode mass of each of the cell types was estimated from their respective bill of materials. The cell energy storage of each of the lithium-ion cell types was assumed to be constant with time as well as across the three scenarios. Using the approach described above, the final input to the MFA model pertaining to number of cells per LIB pack was determined (summarized in Table S3.2 in Appendix A, which distinguishes across scenarios, vehicle types, and cathode chemistries).

Assessment of specific materials comprising each cell within the EV battery waste stream

In the final stage of modeling, the specific materials contained in the battery cells were taken into account. Based on B_t, the total number of waste LIB packs in year t, the percentage of waste LIBs belonging to EV type i in year t ($PE_{i,t}$), the percentage of battery chemistry j in EV battery waste stream (PC_j), and the number of cells per battery pack for EV type i and battery chemistry type j ($D_{i,j}$), the amount of any material y present in the EV battery waste stream for a given year t was estimated as,

$$MO_{y,t} = B_t * \sum_i \sum_j (PE_{i,t} * PC_j * D_{i,j} * m_{y,j}) \qquad (7)$$

$m_{y,j}$ =Mass of a given material y (aluminum, copper, lithium etc.) in a lithium-ion cell of cathode chemistry j

The variable $m_{y,j}$ was obtained from the bill of materials of lithium-ion cells of the four cathode chemistries from the disassembly of 18650 lithium-ion cells (Wang *et al.*, 2014a) and remained constant across the scenarios as well as with time (Table S4.1, Appendix A). The other variables in this part of the model have been discussed in previous sections.

2.2.2 Economic value of materials in EV battery waste stream

The annual value of materials present in the EV battery waste stream was estimated using global spot prices (London Metal Exchange, 2012; Shanghai Metals Market, 2012) and USGS (2012) commodity values of LIB materials (Table S9.1, Appendix A). This estimation only included currently recycled materials (aluminum, cobalt, copper, nickel, steel and iron) as well as high value materials not currently recycled in the U.S. but with high potential for recovery in the future (lithium and manganese) to calculate the "maximum theoretical commodity value" of the EV battery waste stream. The future-oriented characterization of lithium and manganese as high value materials is based on several factors, including current LIB recycling efforts aimed at developing recovery processes for these materials (Paulino *et al.*, 2008; Dunn *et al.*, 2012; Zou *et al.*, 2013; Yang *et al.*, 2013 etc.), limited lithium and manganese resources in the U.S. and the resultant dependence on import of these metals (USGS, 2012), and the potential price rise of these metals with growing demand for EV LIBs. Manganese comprises about 20-25% of a typical lithium-ion cell (Wang *et al.*, 2014a; ANL, 2012) making these cells a viable source for recovery of manganese. Though lithium constitutes only 1-2% of the total cell mass of typical LIBs (Wang *et al.*, 2014a; ANL, 2012), considering an EV battery pack comprising of thousands of cells, the amount of lithium available for recovery would not be negligible. Although lithium carbonate is currently a lower cost input to LIB production (USGS, 2012), the forecast increase in lithium demand by 2020 (Jobin *et al.*, 2009) and potential lag in supply (Kushnir & Sandén, 2012) may trigger lithium price rise in future. In fact, lithium spot prices of about $62/kg have been listed in the Shanghai Metals Market (2012).

Recycling efficiencies of materials and the collection rate of spent EV LIBs were not considered in estimating the commodity value of EV battery waste stream. Other materials in this waste stream that are unlikely to be recycled (graphite, electrolyte, plastics, etc.) were excluded from this valuation. The baseline scenario MFA results were used as basis for these economic estimations.

2.3 Results and Discussion

2.3.1 Estimation of number of EOL battery packs

Based on the parameters defined for each of the three scenarios, the number of EV LIB packs potentially entering the waste stream on an annual basis was estimated (Figure 2.4).

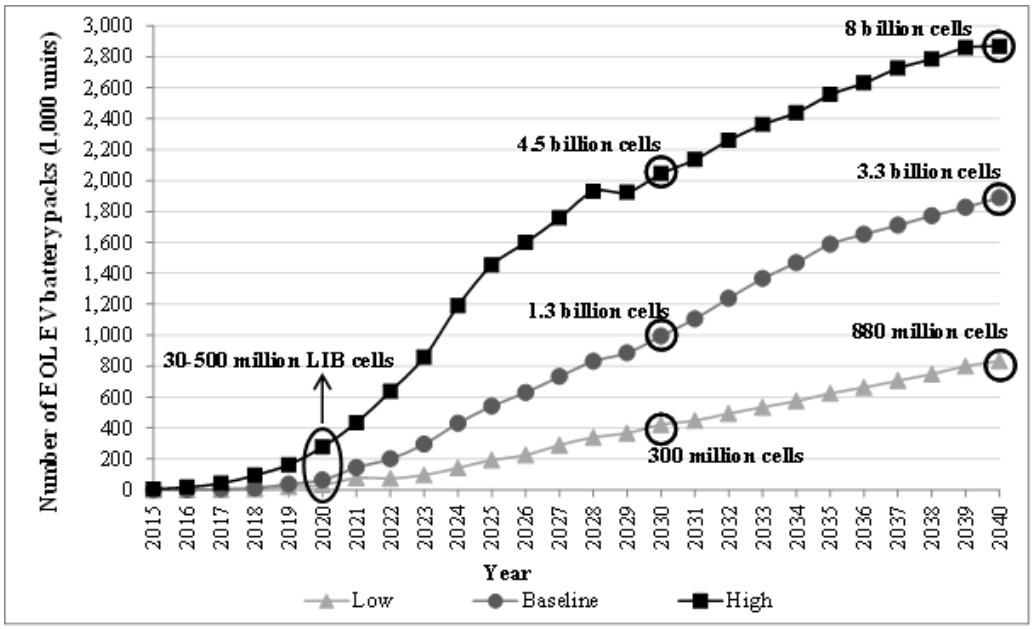


Figure 2.4 End-of-life EV Li-Ion batteries generated annually between 2015 and 2040

While the three scenarios projected similar increase in EV battery waste flows in the U.S. during the first five years of the analysis, results quickly diverge due to differences in input sales and battery lifespans. As per the baseline, approximately 1.9 million LIB packs (each consisting of many cells) could be entering the waste stream annually by year 2040. However, considering the range from the most conservative to most extreme estimates, the waste stream could hypothetically fall anywhere between 0.83 and 2.87 million LIB packs per year by 2040. The cumulative baseline outflow of LIB packs between 2015 and 2040 (21 million packs) was approximately two and a half times greater than the total number of EV battery packs calculated in the "low" scenario (8.7 million packs) and about two times fewer than that of the "high" scenario (40 million packs). Of these LIB packs, between 27-35% would be coming from all-electric and plug-in hybrid electric vehicles, and the remaining 65-73% were estimated to be from hybrid electric vehicles, reflecting the projected sales of each vehicle type.

The battery waste flows were differentiated based on the "Type 1" and "Type 2" classifications of remaining life as described earlier (Table 2.2, also summarized in Figure 2.6 for the baseline). Characterization of EOL batteries into these categories provides some indication of the volume of batteries with the highest potential for suitable reuse applications. For instance, type 2 EOL batteries still hypothetically have remaining EOL life, making them better suited for applications requiring high capacity. In each of the scenarios, Type 2 batteries

represent a sizeable fraction, and despite current hesitance surrounding reuse in vehicles, the number of batteries expected suggests that "re-matching" Type 2 batteries with older vehicles or some other form of cascading use should be studied further.

Scenario	Percentage of Type 1 EOL batteries	Percentage of Type 2 EOL batteries
Low	57%	43%
Baseline	63%	37%
High	62%	38%

Table 2.2 Percentage of Type 1 and Type 2 EOL EV batteries accrued in the waste stream between 2015 and 2040

2.3.2 Lithium-ion cells and attendant material flows in the EV battery waste stream

The EOL EV batteries generated on an annual basis would contain hundreds or even thousands of cells, each consisting of different metals, carbonaceous materials (carbon black and graphite) and other miscellaneous materials such as organic carbonates, lithium salts, binder, plastics, etc. Considering the parameters specified for the baseline scenario, approximately 3.3 billion individual lithium-ion cells may be entering the waste stream annually by 2040. By that point, the cumulative outflows between years 2015 and 2040 would be on the order of 30 billion cells requiring EOL management. The annual waste flows could be as low as 0.88 billion cells (low scenario) or as high as 8 billion cells (high scenario) per year by 2040.

For the baseline scenario in year 2020, the LIB waste stream could contain approximately 3,400 metric tons of lithium-ion cells just from EV application, which is about 4 times the estimated collection volume of waste LIBs from consumer electronics in 2012 (Wang *et al.*, 2014b). In terms of the resultant material flows, the range of scenarios indicate a total EV battery waste stream between 0.33 to 4 million metric tons, with a baseline estimate of 1.3 million metric tons generated cumulatively between 2015 and 2040. Figure 2.5 summarizes annual outflows of battery materials on five-year increments over the long and short term future (Extensive annual estimates of EOL EV LIB material outflows are provided in Appendix A, section S6.3). In this initial estimate, the material-specific composition of that waste stream does not vary, and is also summarized in Figure 2.5.

In comparison with the small body of recent literature on lithium demand for EVs, this MFA predicted relatively conservative outflows, even for the "high" scenario. For instance, Gaines and Nelson (2010) estimated a maximum waste flow of 20,000 metric tons of lithium in 2040 from "optimistic" EV deployment, a prediction about 10 times greater than our baseline

scenario estimates for that year. To put our estimates in a global context, this study forecast cumulative lithium outflows between 2020 and 2040 between 4.5 thousand to 55 thousand metric tons for the U.S. On a global basis, Gruber *et al.* (2011) estimated 860 thousand metric tons of potentially recoverable lithium from EV batteries in the same time frame (with 100% recycling participation and 100% lithium recovery).

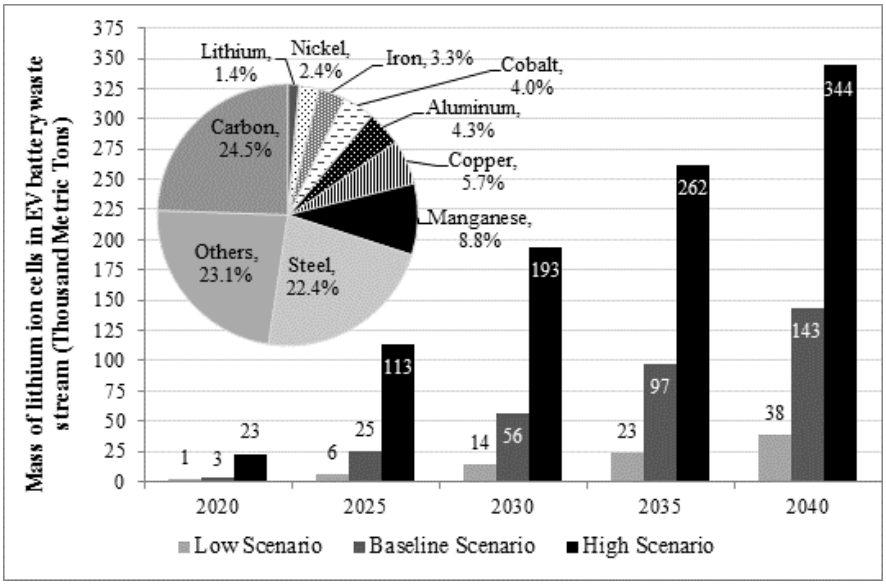


Figure 2.5 Mass of Li-Ion cells in EV battery waste stream (In the pie-chart, "carbon" includes carbon black and graphite. "Others" include plastics, binders, electrolytes and other non-metals like phosphorus etc.)

The disparity observed between the low and high scenario in this chapter is indicative of the variability in estimates of EV sales, the battery lifespan (and resultant need for replacement battery packs, particularly in the high scenario) as well as the parameters determining the number of cells per EV battery pack. Even with these uncertainties, one can begin to analyze results further, using the baseline scenario as a focal point (to minimize the amount of data presented in the main text). The baseline scenario was further characterized on the basis of battery inputs, outputs, and material characteristics. Figure 2.6 summarizes these characteristics for the cumulative input of LIBs in electric vehicles between 2009 and 2034, and the net EOL battery outflows between 2015 and 2040.

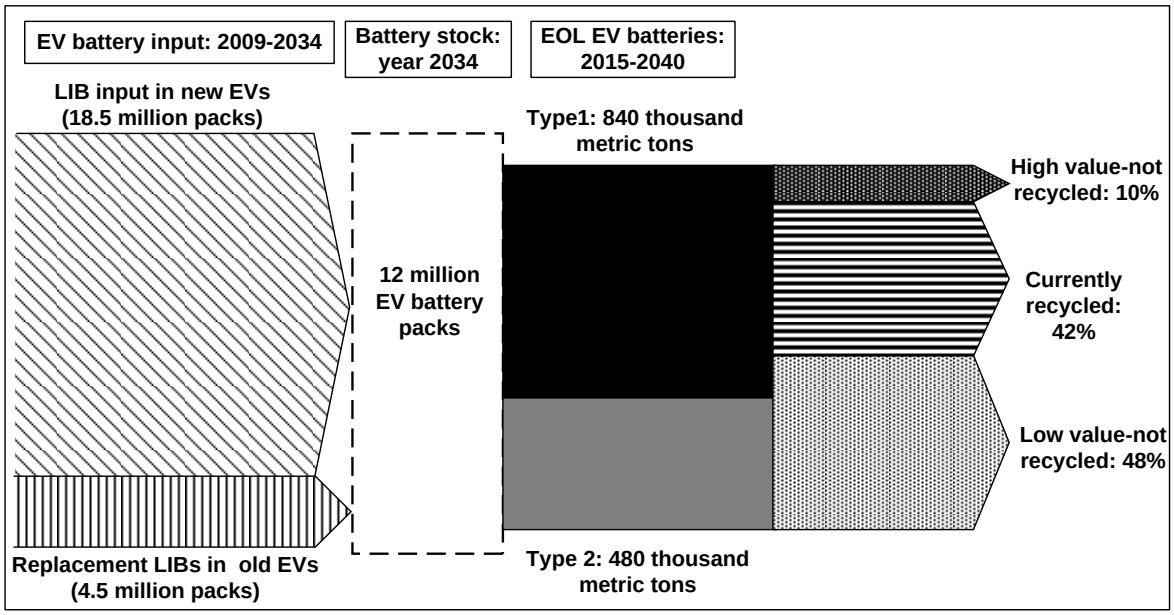


Figure 2.6 EV battery material inflow and outflows-Baseline Scenario. Thickness of each bar corresponds to the relative mass of material in each category.

The majority (80%) of new batteries entering use would be paired with new EVs sold in the market, while the remaining 20% would be replacement batteries for existing in-use EVs (Figure 2.7). About 63% of the batteries leaving use were "Type 1," with no remaining life for EV applications; while the remaining 37% "Type 2" batteries may have been discarded before their true EV end-of-life. Previously shown materials analysis (Figure 2.5) distinguishes different materials contained in the battery waste stream, but the ultimate fate of these materials depends on whether an infrastructure and market exist for their recycling back into productive use. The potential of each material to be recycled was determined by assessing current recycling practices and secondary markets available for these materials (USGS 2012). Based on the potential to be recycled, the materials expected in the EV battery waste stream were categorized as currently recycled, high value-not recycled, and low value-not recycled materials.

Of the estimated battery outflows, low value materials, which are currently not being recycled and are not expected to be in the future, could constitute 48% of the EV battery waste stream and include constituents such as graphite, carbon black, lithium hexafluorophosphate ($LiPF_6$), organic carbonates (such as ethylene carbonate or dimethyl carbonate), binder (polyvinylidene fluoride) and mixed plastics (polypropylene, polyethylene). Apart from plastics, none of these materials have a secondary market at present and it would not be economically viable to recover them from the waste stream. Moreover, as a mixed grade of plastics would be

present in the battery waste stream, their recovery would not be likely due to high contamination. Because these materials are not suitable for recycling, infrastructure must be equipped to accommodate their introduction to landfills or other disposal routes. As such, relevant environmental and health impacts should be anticipated. The carbonaceous material present in the EV battery waste stream could raise concern in the future owing to their large quantity in the waste stream (Figure 2.5) and knowledge of potential health impacts of particulate carbon (e.g., exposure to graphite dust can adversely affect respiratory system and pulmonary function (NIOSH, 2007)). The electrolyte chemicals present in LIBs can have toxicity concerns as well. For instance, the electrolyte salt $LiPF_6$ is a hygroscopic substance and in presence of moist air or water forms hydrogen fluoride gas (Archuleta, 1995), which has severe environmental risks and toxicity concerns (EPA, n.d.). Similarly, organic carbonates used as electrolyte solvents are mildly toxic, volatile and flammable compounds, producing toxic fumes on decomposition (Vimmerstedt *et al.*, 1995). Environmental impacts of EV battery waste could also be a concern due to the non-biodegradability of binder and other plastics in lithium-ion cells.

Another 42% of the materials in the cumulative EV battery waste stream would include materials that are currently and expected to continue being recycled according to statistics from the USGS (2012). This fraction includes metals such as aluminum (57,000 metric tons), cobalt (52,000 metric tons), copper (75,000 metric tons), nickel (32,000 metric tons), steel (295,000 metric tons) and iron (43,300 metric tons). These material masses in waste stream are on a cumulative basis estimated between years 2015 and 2040. The remaining 10% of the EV battery waste stream would include two high value materials that are currently not recycled in the U.S., i.e., lithium (18,000 metric tons) and manganese (116,000 metric tons). Many of these metals (lithium, aluminum, nickel and cobalt) have high embodied energy when extracted from virgin resources (ecoinvent Centre, 2007). Hence, recycling of LIBs offers a dual benefit: avoided energy inputs for production of primary metals and potential economic revenue from material recovery, which is particularly high for cathode chemistries like Li_2CO_3 and NCM that contain 10-17% by weight of high-value cobalt.

2.3.3 Economic value of materials in EV battery waste stream

Considering the baseline scenario with a mix of lithium-ion cell chemistries, the total EV LIB waste stream may contain materials valued at approximately 3.8 billion USD on a

cumulative basis between 2015 and 2040. This estimate is the maximum theoretical commodity value of the EV battery waste stream considering the potential value for materials that currently have recycling infrastructure in the U.S, and does not take into account material losses that would occur due to recycling inefficiencies. The total possible waste stream value would be increased by over 1.5 billion USD if Li and Mn are also included.

The actual economic value of the EV battery waste stream would depend on the LIB collection rates, the recovery rates of the various materials present in the stream, and the cost of recycling itself. Considering recent recycling efficiencies (see Appendix A, Table S10.1), commodity value of approximately 3 billion USD could be obtained between 2015 and 2040 by recovery of metals such as aluminum, copper, nickel, cobalt, iron and steel assuming that 100% of batteries in the waste stream can be collected for recycling. Wang *et al.* (2014b) analyzed the profitability of LIB recycling facilities for several possible future co-mingled LIB waste streams based on the current recycling efficiency of materials in LIBs: the potential value from recycling one metric ton of LIBs ranged from \$860 for $LiMn_2O_4$ cathode batteries to \$8,900 for $LiCoO_2$ cathode batteries. Continued development of advanced separation processes could increase the recycling efficiencies of materials present in EV LIBs and hence the economic motivation for recovering materials from these batteries. For example, a 10% improvement over current recycling efficiency for cobalt could raise the recycling revenue by 9% for cobalt based LIBs while a 10% improvement in copper recycling efficiency would only improve revenue from LIB recycling by 1% to 5%, depending on the cathode chemistry (Wang *et al.*, 2014b).

The materials potentially recoverable by EV LIB recycling could be used as inputs to the parent battery industry, as this sector is predicted to become more resource intensive as vehicle deployment increases. Increasing availability of secondary material sources would reduce U.S. dependency on foreign resources in the long run. Gaines and Nelson (2010) estimated that recycling LIBs could meet almost 50% of the lithium required for battery production in the U.S. by 2040. However, the recyclability of the EV battery waste stream and hence, the economic gains from battery recycling is likely to depend on the battery technology prevalent in the future in terms of cathode chemistry as well as the form factor of the lithium-ion cells used in these batteries. At present there is significant uncertainty in this domain, which is analyzed in the subsequent sections, along with the uncertainty due to differences in MFA parameters such as EV sales, battery lifespan and number of cells per EV battery pack.

2.3.4 Uncertainty Analysis

EV sales and battery lifespan: It was established by the range of scenario results that the volume of LIBs in the EV battery waste stream would be highly dependent on EV sales and the actual battery lifespan. The sales of electric vehicles will depend on a number of factors in the mid- and long-term, such as oil prices, battery and vehicle cost, EV and battery technology, government subsidies, policies and regulations etc. (EIA, 2012). The lifespan of lithium-ion batteries in EV application will depend on battery technology progress as well as usage patterns at the consumer level. Long battery lifespans would have a two-fold benefit, first reducing the need for a second (or even third) replacement battery and thereby reducing the cost of ownership of electric vehicles, and second, raising the potential for post-EV battery reuse, which can also defray costs across the battery life cycle.

Uncertainty analysis was performed to tease apart the role of sales and lifespan parameters on LIB waste flows, by holding one parameter constant and varying the other (Appendix A section S7.1). When the EV sales estimates are held constant at the baseline level, cumulative (2015-2040) LIB outflows increased 16% or decreased 15% from the shortest lifespan ("high" scenario) to the longest ("low" scenario). On the other hand, when the baseline EV LIB lifespan distribution was combined with the high and low scenario EV sales figures, cumulative (2015 to 2040) outflows of EOL packs could increase by as much as 62% and decrease by 52%, respectively. It is evident that even though the battery lifespan distribution plays a role in influencing the volume of EV battery waste stream, electric vehicle sales will be the governing factor influencing EV LIB waste flows in the future.

When a longer EV lifespan of 16 years was tested for model sensitivity, the cumulative (2015 to 2040) outflows of these batteries into the waste stream changed by less than 2%, although the annual waste stream volumes varied, as shown in Appendix A, section S7.3. Further, the percentage of Type 1 and Type 2 EV batteries also changed with increasing lifespan of EVs. For instance, when a longer EV lifespan of 16 years was assumed for the baseline scenario, the percentage of Type 2 EV batteries estimated to accrue in the waste stream between 2015 and 2040 reduced from 37% to 23% percent (Detailed analysis in Appendix A section S7.3).

Cell and battery energy storage and battery pack components

The energy storage of the battery pack also plays an important role in determining the amount of cells per pack, and thus the materials present in the EV battery waste stream. A sensitivity analysis was conducted wherein the baseline cumulative outflow of EOL EV battery packs was held constant while the estimated EV battery pack energy assumptions were varied between the low and high scenarios (Appendix A, section S7.2). The resulting estimates of total material mass of the EV battery waste stream decreased by up to 29% or increased by up to 40% when the low or high scenario cells per pack assumptions were applied, as compared to baseline flows.

Hence, it follows that battery and EV technology (in terms of electric miles and vehicle mileage) would play a major role in governing flows of EV batteries in the waste stream. Throughout the EV sales forecast timeline considered in the model (2009 to 2034), the same battery chemistries were assumed and the energy storage by individual cells was held constant. According to Srinivasan (2008), the energy density of lithium-ion batteries has been increasing at the rate of approximately 5% per year over the last one and a half decade. The average energy density of a typical 18650 cell is approximately 200 Wh/kg (Howard & Spotnitz, 2007). According to Srinivasan and Lipp (2003), when lithium-ion batteries were introduced in the early 1990s, this number was around 90 Wh/kg. In other words, technological innovation has doubled the energy density of these batteries. It is expected that this trend will continue in the future, with ongoing research and development to introduce nano-materials and mixed-metal technology for higher energy density (Ritchie & Howard, 2006; Howard & Spotnitz, 2007). As this technology advances, fewer cells per pack and/or less material per cell may be achievable, which may reduce total material flows to the EV battery waste stream.

The mass and composition of the EV battery waste stream modeled here has considered only the cells within EV battery packs. For a typical Tesla Roadster EV battery, with 6,800 cells weighing approximately 46 grams each, the total cell weight is about 313 kg, but the entire battery mass is about 450 kg (Berdichevsky *et al.*, 2006). The difference in weight is attributed to the battery pack casing, module components, electronic parts, thermal insulation, etc. (Dunn *et al*, 2012), which can account for anywhere between 10-30% of the EV battery weight. Considering this entire battery system, the actual material flow entering the waste stream would be even greater than estimates calculated here. However, including other EV battery pack

components would require further modeling, as these components may have much higher reuse potential than the batteries themselves and may not enter the waste stream at the same time as the LIB cells (Cready *et al.*, 2003).

Battery chemistry and form factors

All results shown to this point have followed an assumed mixture of different cathode chemistries, and are based only on 18650 (cylindrical) form factors. In the future, composition of the EV battery waste stream will depend heavily on the actual cathode chemistries and form factors selected by auto manufacturers. For example, new EV models such as the GM Volt and Nissan Leaf employ prismatic cells, while the 18650 form factor continues to be used in the Tesla roadster BEVs. Though this MFA model enables a reasonable approximation of the material and economic flows of the EV battery waste stream, there is a need to assess the role that a single dominant chemistry and prismatic form factor could play in determining the volume, composition, economic value, and recycling potential of this waste stream. Using the baseline scenario estimation for EOL LIB packs, the number of cells and material mass were estimated for the following cases:

(A) Scenarios in which a single dominant lithium-ion cathode chemistry (18650 form-factor) would be employed in all EV batteries. The four candidate chemistries were Li_2CO_3 (LCO), $LiMn_2O_4$ (LMO), $LiFePO_4$ (LFP) and mixed metal (NCM), each modeled individually as a dominant cathode chemistry.

(B) A scenario in which prismatic cells instead of 18650 cells were used. The chemistry mix of EV battery waste stream was assumed to be same as in the 18650 scenario (10% LCO, 30% each of LMO, LFP and NCM cells), however here only the LCO cells were of 18650 form factor (consistent with adoption by some auto makers, like Tesla) while the remaining 90% of the LIBs consisted of prismatic cells. See Appendix A for scenario details.

When compared to the baseline EV LIB outflows (a mix of potential chemistries in 18650 cells), the number of lithium-ion cells in the waste stream was roughly the same for the different dominant chemistry scenarios. The LFP scenario resulted in the highest number of waste cells, approximately 35 billion, cumulatively between 2015 and 2040 (as compared to the baseline 30 billion cells). When considering prismatic cells, results showed an interesting

dichotomy: the total number of cells in the EV battery waste stream would reduce drastically if most EVs employed prismatic cells- to almost 4.4 billion cells in the waste stream between 2015 and 2040, but the net flow and type of materials into the waste stream would remain relatively constant. This consistency held across all different scenarios of cathode chemistry and form factor, which ranged between 1 and 1.5 million metric tons of battery waste on a cumulative basis between 2015 and 2040 (Figure 2.7). Further, the recyclability of the EV battery waste stream would also vary with the battery chemistry and form factor (Figure 2.7). For instance, if the $LiMn_2O_4$ chemistry is predominantly used, then the EV battery waste stream would contain negligible amount of currently recyclable materials and large quantities of low value materials that are not recycled. However, its recycling can generate value if the currently non-recycled high value materials like lithium and manganese can be recovered (Figure 2.8). Similarly, even though both the form factor scenarios (Chemistry mix "Base Case" and 90% prismatic) consisted of the same distribution of LIBs belonging to the four cathode chemistries in the waste stream, the fraction of recyclable materials is slightly higher in the case of cylindrical cells, which require more metallic casing components (typically aluminum or steel).

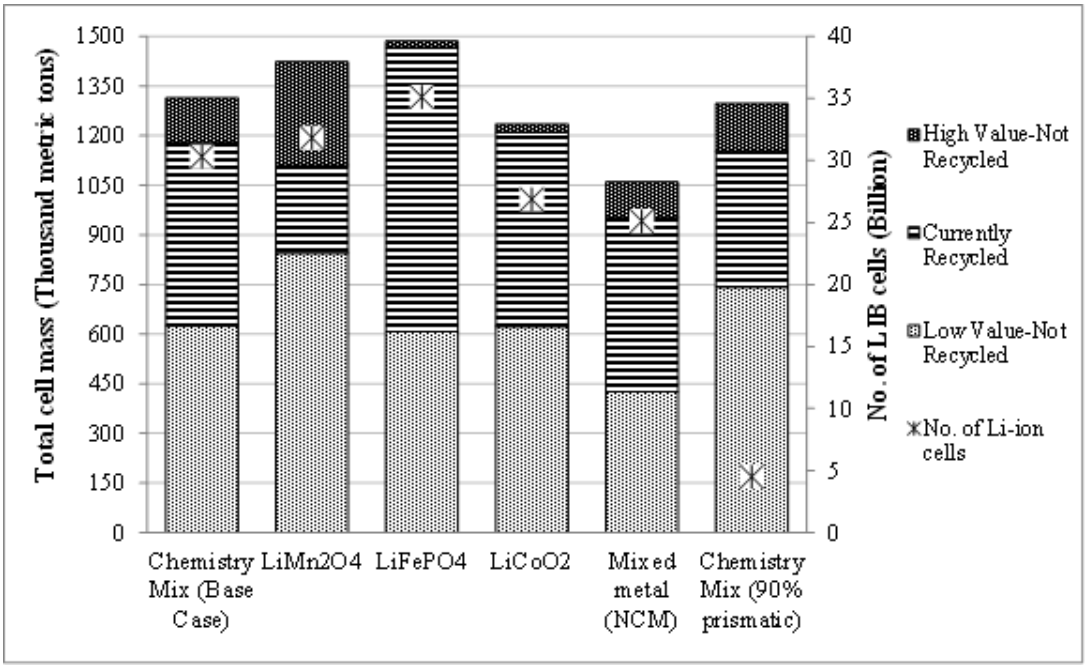


Figure 2.7 Recyclability of EV battery waste stream under different scenarios of Li-Ion cell cathode chemistry and form factor (cumulative flows from 2015 to 2040)

An important caveat to these findings is that the high percentage by weight of recycled materials in the EV battery waste stream may not translate into high economic gains from LIB

recycling. For instance, even though 58% of a $LiFePO_4$ cell waste stream consists of currently recycled materials, the relative economic value of this stream is lower than any other chemistry (Figure 2.8). While the relative volumes of recycled materials are higher in the case of all cylindrical cells, as compared to the prismatic form, there is no significant difference in the total commodity value of materials in the waste stream associated with these two scenarios.

In fact, the economic feasibility of EV lithium-ion battery recycling in the future would not only depend on collection and recycling efficiencies, but also on the chemistries selected for EV battery manufacturing and ultimately ending up in the waste stream. A large scale use of LCO and NCM chemistries for EV batteries would translate into high economic values of the EV battery waste. In the LCO and NCM chemistry scenarios, currently recycled materials would constitute about 50% of the materials in the battery waste stream by mass, but could account for 86% of the economic value of that stream. However, as battery manufacturers shift to cheaper chemistries such as LMO and LFP, to improve performance and avoid high cost and scarcity of cobalt resources (Nishi, 2001), the resulting value of the currently recycled portion of the waste stream could be reduced to as low as 340 million USD, on a cumulative basis (2015-2040).

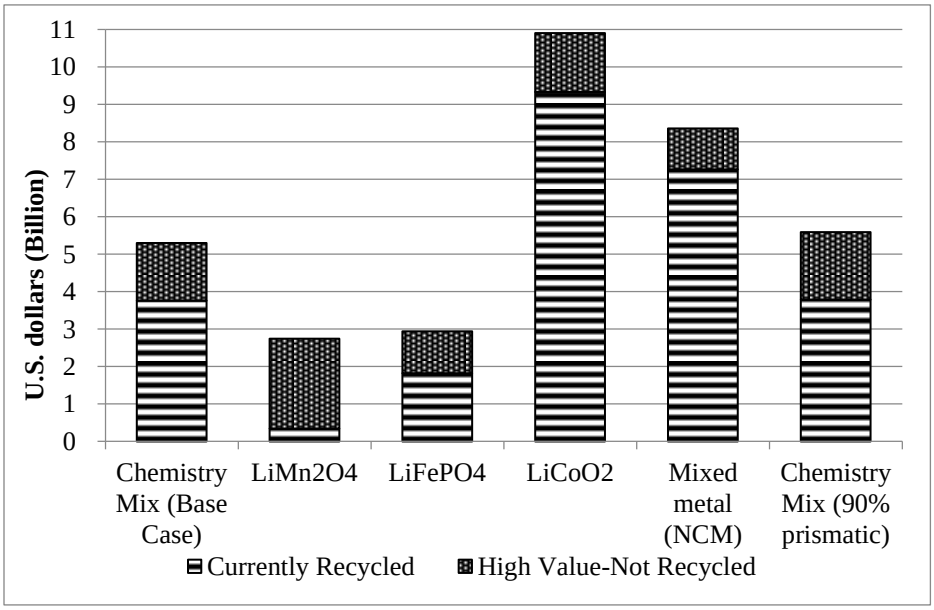


Figure 2.8 Cumulative material value of EV battery waste stream (2015-2040)

Cost efficient recycling procedures for the recovery of high value materials, like lithium and manganese, which are currently not recycled in the U.S. would add some incentive towards recycling of economically unattractive LIB chemistries. In the LMO chemistry scenario,

currently non-recycled high value materials constitute 22% of the waste flows by mass, but account for 87% by value. Even more extreme, in the LFP battery chemistry scenario, the currently non-recycled high value materials accounted for only 1% of the waste EV battery cells by mass, but could make up to 38% of the total material value. Although Wang *et al.* (2014b) conclude that for a LIB recycling facility to be profitable, the proportion of LiCoO2 cathode batteries in the waste stream needs to be 21%, improved recycling processes in the future is expected to improve the overall profitability of recycling EOL LIBs.

2.4 Conclusions

It is clear that EV batteries will emerge as a future waste management challenge, with projected annual waste flows reaching as high as 340,000 metric tons by 2040. Because of the high volume, complexity and variety of materials forecast in the EV battery waste stream, it is evident that multiple waste management routes must be developed for EOL LIBs from electric vehicles:

1) reuse avenues for battery cells and packs with remaining life,
2) recycling infrastructure capable of recovering high value material from multiple battery chemistries, and
3) safe disposal routes for materials with minimal or no secondary value or recovery infrastructure.

Results also indicate that high variability in the potential economic value associated with the projected LIB waste stream may pose challenges for development of recycling infrastructure. At present, profit from LIB recycling is constrained by high collection and processing costs (Wang et al., 2014b). Currently, there is no federal regulation that mandates LIB recycling, and only two states - California and New York - have passed regulations banning landfill of these batteries. To overcome potential economic constraints of LIB recycling, particularly for less valuable, non-cobalt chemistries, the recovery process and infrastructure may require policy intervention to reach economies of scale.

Apart from economics, environmental health and safety may also motivate policy attention to future EOL LIB management. The absence of consistent infrastructure and regulations for lithium-ion battery recycling may increase the potential risk of environmental impact due to EOL EV batteries. Though the state of California classifies them as hazardous due

to the presence of cobalt, LIB wastes are included under EPA's Universal Waste Rule (Gaines and Cuenca, 2000) and are in general not considered to be hazardous for the environment due to absence of toxic elements like lead, mercury or cadmium. However, landfill of EV LIBs may introduce environmental risks due to leakage of organic electrolytes, presence of heavy metals such as copper and nickel (Shin *et al.*, 2005), reactive lithium salts, and large quantity of carbonaceous materials (graphite and carbon black).

A number of uncertainties still exist, and exact estimation of future waste flows will depend on the ability to further refine the forecasts of EV sales, battery and EV lifespan, and trajectories of battery technology deployment. Waiting until such refinements are possible, though, presents a risk of not allowing sufficient time for domestic infrastructure and policies to react to the emergence of a full scale battery waste stream. Thus, proactive advancement of a robust EOL battery reuse, recycling, and disposal system will be required to handle the variety and volume of materials expected. Moreover, the MFA model provided here can be adapted to extend the analysis of LIB wastes as more definitive data become available.

CHAPTER 3: ENVIRONMENTAL TRADE-OFFS ACROSS CASCADING LITHIUM-ION BATTERY LIFE CYCLES

3.1 Introduction

While electric vehicles have the potential to reduce greenhouse gas emissions from the transport sector, a major obstacle towards their large scale adoption has been the high cost of lithium-ion batteries (Axsen et al., 2010). Recent literature and technical analysis estimate that at the end of their service life in EVs, LIBs would still have 80% of their original capacity intact (Hoffart, 2008; Cready et al., 2003; Neubauer & Pesaran; 2011; Marano et al., 2009). Though insufficient for automotive application, they can be reused in stationary applications which have a much lower capacity limit and hence, are less demanding applications (Hein et al., 2012). As indicated from the results in Chapter 2, as high as 43% of the future EV LIB waste stream can have reuse capability in stationary use, hence immense potential exists in utilizing these battery systems for small and large scale stationary energy storage applications. This cascaded reuse of EV LIBs would enable original equipment manufacturers (OEMs) to distribute the high initial cost of the EV battery over two lifespans. Several studies confirm the economic feasibility of EV LIB reuse for stationary energy storage applications like intermittent renewable storage, grid support, and power back-up (Heymans et al., 2014, Williams and Lipman, 2010; Neubauer et al., 2012; Neubauer & Pesaran; 2011; Cready et al., 2003, Beer et al., 2012, Narula et al., 2011 etc.). As a result, stakeholders in both the EV and the utility sectors have interest in pursuing EV battery reuse.

In the EV sector, successful battery reuse applications can potentially reduce the net life cycle costs of EV and LIB systems, and by extension, the battery cost borne by EV customers, thus impacting consumer purchase decisions and battery usage patterns (Neubauer et al., 2012; Williams and Lipman, 2010). Opening new reuse pathways may also mitigate rising disposal and compliance concerns over the growing battery waste stream as forecasted in Chapter 2 (Richa et al. 2014). For example, California and New York state legislators have issued disposal bans on rechargeable batteries (including but not limited to LIBs) in their states (CA Code, 2006; New York State Rechargeable Battery Law, 2010) restricting their disposal as solid wastes. As a result, OEMs now face disposal costs for these cells. Although these costs can be offset by recycling revenue, the materials contained in EV LIBs have been evolving towards less-

expensive cathode chemistries, which is expected to reduce the overall economic incentive for recycling under current conditions (Wang et al., 2014a; Wang et al., 2014b).

Similarly, studies suggest that reuse of EV LIBs in stationary applications would provide cost-efficient services to the stationary energy storage markets, improve utility operation and provide a low-cost revenue source for this sector (William and Lipman, 2010; Beer et al., 2012). Battery energy storage has a considerable demand for grid as well as off-grid applications. These applications range from power backup, load following, renewable firming, peak shaving, energy arbitrage, etc. (Cready et al., 2003; Albright et al., 2012; Soloveichik, 2011). Over the years, a range of battery technologies have been tested as well as applied for these utility applications. These applications can be broadly categorized into two (Doughty et al., 2010):

(a) Energy applications that involve long charging sessions, followed by discharging the battery over several hours. Typically, such application would require a single charge-discharge cycle of the battery in a day.

(b) Power based applications that involve short charging periods and discharges of few seconds to minutes. Such applications may require the battery to be charged or discharged several times a day.

The major criteria for selecting battery systems for these utility applications have been costs, reliability, performance, and battery design. While a range of battery technologies have been developed and tested for utility sector applications, only few such as lead acid (PbA), nickel-cadmium, sodium-sulfur, and vanadium-redox flow batteries have been commercially adopted due to their favorable cost and reliability factors (Doughty et al., 2010). However, among these technologies, PbA batteries (both flooded and valve regulated) are the most widely used for different types of energy storage applications (Soloveichik, 2011). In future, cascaded use of retired EV LIBs in such applications can prove to be competitive with PbA battery technology.

According to Beer et al. (2012), retired EV LIBs are expected to have higher capacity and cycle life expectancy than incumbent PbA batteries in stationary applications. This may not hold true in all cases, as the reuse potential of LIBs would depend on the battery itself, particularly the cathode chemistry (Burke, 2009) and its condition post-EV use, which can be highly variable depending on vehicle usage cycles and temperature conditions to which LIBs are subjected during EV use (Cready et al., 2003). Currently, the remaining and degrading capacity and the

declining efficiency of used batteries in secondary application are relatively unknown (Ahmadi et al., 2014b). If refurbished EV batteries provide a considerably low level of functionality or service life, the probability of the cascading reuse route may decrease.

In addition to these economic incentives, environmental benefits could also be realized due to cascading reuse of EV batteries in secondary applications. By extending EV battery lifespan through reuse, LIB and EV producers can conceivably reduce the net environmental impact of the battery pack attributed to the EV itself. Similarly, by utilizing used vehicle batteries, energy utility providers can avoid the impacts associated with producing and using a new PbA battery. The degree of benefit would, however, depend on the reliability of reused EV batteries, which will ultimately impact the reuse decision by stakeholders, as well as methodological choices in the approach to allocating battery impacts between the first and second lives. To obtain a better understanding of the feasibility issues of EV LIB reuse, life cycle assessment (LCA) can be applied to estimate such environmental tradeoffs relevant to each stakeholder's perspective across a wide variety of technical and methodological parameters.

Several LCA studies have analyzed environmental impacts of LIBs as part of a broader effort to understand tradeoffs in the EV life cycle (Majeau-Bettez *et al*., 2011; Notter *et al*., 2010; Zackrisson *et al*., 2010; Samaras and Meisterling, 2008; Ishihara et al., 2002; Hawkins et al., 2013; Matheys et al., 2009, Ellingsen et al., 2013). These studies are primarily restricted to the production and use phase of LIBs. Due to a lack of publicly available and transparent data, only in rare cases has recycling of EV LIBs for constituent metals been considered to be part of the LCA scope (e.g. Ishihara et al., 2002; Matheys et al., 2009; Dunn et al., 2012). Considering that battery reuse post-EV application could be a common practice in future (Howell, 2012), it is essential to include a consideration of secondary usage in LCA studies to provide a more comprehensive understanding of battery and EV environmental impacts.

A few recent LCA studies by Ahmadi et al. (2014a), Cicconi et al. (2012) and Genikomsakis et al. (2013) demonstrated a net reduction in CO_2 emissions can be achieved by reusing EV LIBs, particularly due to their utility for smart grid or renewable energy applications and the attendant reduction of fossil fuel use. Building upon these LCAs, this study analyzes the environmental performance of post-EV LIBs against basic feasibility criteria for reuse (e.g. battery lifespan in secondary application), thus attempting to provide an enhanced perspective of environmental implications of battery reuse. Given the interrelated objectives of both the EV and

the utility sectors, this LCA was framed in dual ways to address both perspectives individually, so as to avoid double counting the potential benefits or impacts and to provide scenario-specific findings relevant to decisions being made within each sector:

1. From the perspective of EV and LIB producers, to what extent can the addition of a secondary reuse application minimize the net environmental impacts across the battery life cycle (the "extended life cycle" case)?
2. From the perspective of the utility, how does the environmental impact of a post-EV, used LIB compare to that of a new PbA battery, particularly given unknowns in reliability of used LIB (the "stationary energy storage" case)?

To address these questions with LCA requires concurrent resolution of the methodological challenges surrounding allocation of environmental impacts of LIB production and end-of-life (EOL) management between EV and stationary use battery. While this challenge can be avoided for the first case by system expansion to account for the avoided PbA system, a direct comparison of cascaded LIBs to incumbent PbA batteries requires allocation choices for upstream LIB impacts. The EV and stationary use battery life cycles together represent a "cascaded use multi-functional system" where LIB cells and module components are passed on from EV application to stationary application, and as such part of the environmental burdens from the upstream can be considered to be transferred to the downstream battery system in the cascade thus creating a need to allocate impacts of a set of common activities to the stationary energy storage system. These allocation decisions are further complicated by system attributes, such as the change in function and use of the battery between life cycles, the lack of an organized recycling network for EV LIBs (Hoyer, 2011), and the immense uncertainty surrounding the roles and business models to be adopted by each stakeholder group in EV LIB recycling (Idjis et al., 2013).

The focus of most studies dealing with allocation problem in LCA of multi-functional systems has been material cascades or "open-loop recycling" of materials (e.g. Ekvall and Tillman, 1997; Azapagic and Clift, 1999; Boguski et al., 1994; Klöpffer et al., 1996; Kim et al., 1997; Newell and Field, 1999; Rydberg, 1995, etc.).Very few studies have explored allocation in LCA of product cascaded reuse, such as Werner et al. (2007) and Streicher-Porte et al. (2009). The choice of allocation methodology employed can have a decisive impact on results of LCA

studies (Rydberg, 1995; 1998; Streicher-Porte et al., 2009). Therefore, one additional objective of this study is to determine the extent to which allocation choices across the product cascade influence the decisions resulting from analysis of the two cases.

3.2 Method

While much of the modeling and data analysis is the same for the two cases analyzed, each is described independently here, and points of methodological divergence are highlighted. To reflect the two stakeholder perspectives, two goals and scopes were specified to analyze environmental issues surrounding cascading reuse for 1) the EV sector (to analyze environmental implications of extending the life of an EV LIB pack with a cascading second use phase), and 2) the utility sector (to compare the environmental impacts of a stationary energy storage system based on refurbished EV LIBs with an equivalent PbA battery based system). The two goals are described in turn.

3.2.1 Goal 1: To analyze environmental implications of extending the life of an EV LIB pack with a cascading second use phase:

For the first objective, an extended life cycle model has been adopted, wherein all stages in the life cycle of an EV battery are taken into consideration (Fig. 3.1). The primary objective is to determine if, from the perspective of the OEM, cascading reuse of the LIB can create a significant reduction in the net EV LIB life cycle environmental impacts. Here, the system is expanded to include the "avoided product system" for a PbA battery that would provide equivalent functionality in the energy storage system. The PbA battery is modeled here as the avoided system because it is a widely used incumbent technology for stationary and industrial purposes due to its affordability (Soloveichik, 2011).

Functional unit - Case 1: The functional unit for this system is a 24 kWh lithium manganese oxide ($LiMn_2O_4$) battery pack for a battery EV (BEV) weighing 223 kg and giving 100,000 miles operation during the EV lifetime; the cells from which are subsequently used in stationary energy storage. This mileage corresponds to an 8 year service life, based on similar warranty terms for the Nissan Leaf BEV battery (Nissan North America, Inc., 2014). The choice of $LiMn_2O_4$ cathode chemistry was due to its current use in Nissan Leaf and GM Volt cars (Lu et

al., 2013), which are the largest selling all-electric and plug-in hybrid electric vehicles, respectively in the United States (Inside EVs, 2015). While there is no common consensus as to which LIB chemistry would dominate the future EV market, several studies have suggested that owing to high cost and safety concerns of cobalt batteries, EV manufacturers could replace cobalt based LIBs with less expensive technologies (Ritchie and Howard, 2006; Wang et al., 2014b; Nishi, 2001). $LiMn_2O_4$ cathode chemistry is one of the likely candidates due to low cost and high availability of manganese (Wellbeloved et al., 1990). The EV battery pack was modeled using the Argonne National Laboratory [ANL] BatPac model (ANL, 2011) by drawing parallels with the existing design of a Nissan Leaf battery pack (Blanco, 2010) as the reference for a BEV battery pack design, which was comprised of 192 prismatic form factor cells grouped into 48 modules.

System boundary- Case 1: The system modeled includes LIB production, LIB use during the EV life, battery refurbishment, secondary use in stationary energy storage, recycling of batteries, as well as the avoided life cycle of a PbA battery system for stationary energy storage (Fig. 3.1). In this case, allocation has been avoided, by system expansion to include the eliminated demand for a PbA battery.

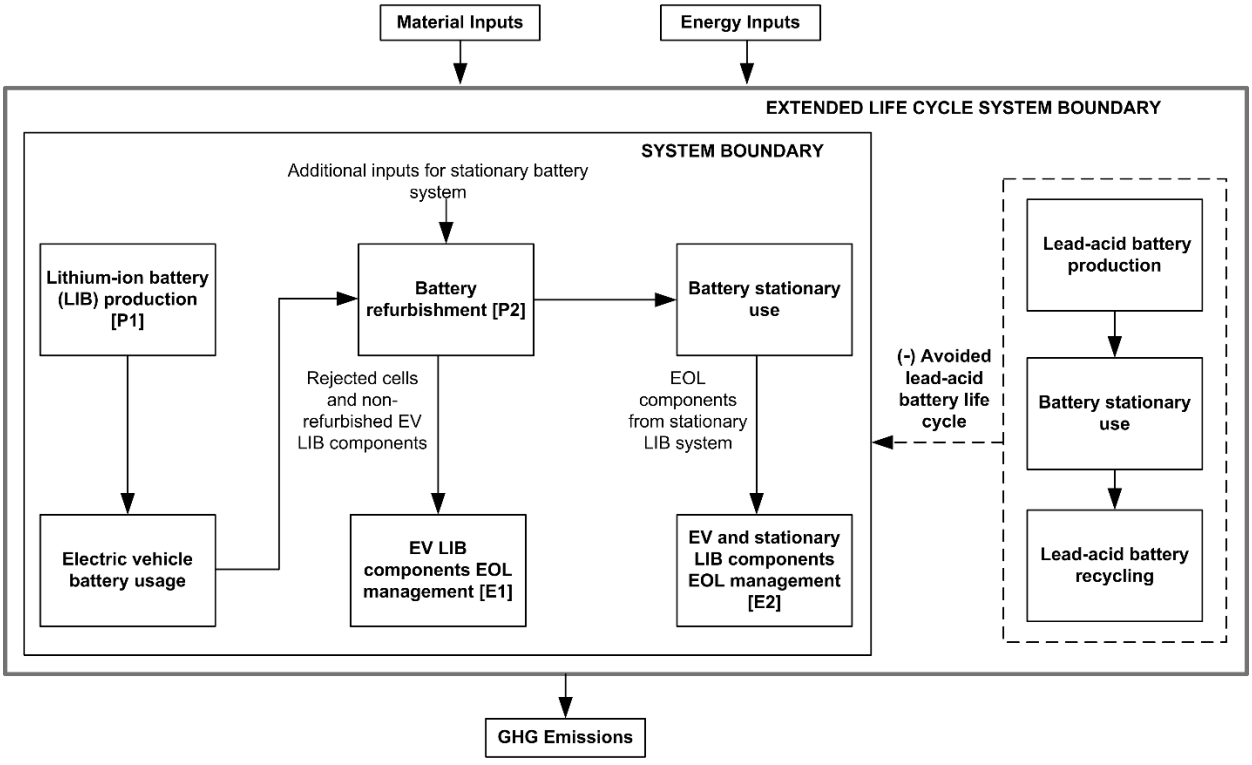


Figure 3.1 System boundary to assess environmental implications of EV LIB with a cascading second use phase in its life cycle (case 1: extended life cycle). The dotted lines represent the extended system boundary that provides

credits for avoiding the life cycle of an equivalent functionality PbA battery. This system corresponds to results shown in Figs. 3.3 and 3.4

3.2.2 Goal 2: To compare the environmental impacts of a stationary energy storage system based on refurbished EV LIBs with an equivalent PbA battery based system.

For the second objective of this LCA, the refurbished EV LIB based energy storage system is considered independently, from the perspective of a utility operator facing the choice of either using a refurbished LIB or a new PbA battery system. Thus, an equivalent functionality PbA battery based stationary energy storage system is used as a basis for comparison.

Functional Unit - Case 2: The functional unit for this goal is defined as a stationary energy storage system, delivering 150 kWh of energy on a daily basis for 20 years. This definition is based on a similar assumption by Rydh (1999) for a battery system providing electricity requirements for several remote houses for 10–70 persons. To translate this functional unit into a specific battery case, the expected depth of discharge (DoD) (the percentage of the battery capacity that is discharged while delivering energy) was taken into account. DOD is restricted to preserve battery life, minimize capacity fade, and ensure safe operation (Axsen et al., 2008, Peterson et al. 2010). For stationary battery operation, DoD is assumed to be between 33-42%, which is capable of providing a 5-year lifespan for PbA battery (Rydh and Sanden, 2005; Bindner et al., 2005), while representing a safe operating range for second use LIBs. This DoD assumption results in an extreme sizing scenario wherein both stationary battery systems are sized equally at 450 kWh while delivering only a fraction of stored energy, i.e. 150 kWh (Rydh, 1999; Rydh and Sanden, 2005) (See Appendix B, section S4). For a refurbished LIB-based system, meeting this functional unit would call for 4,500 post-EV cells with a combined energy storage capacity of about 450 kWh. Such a system would weigh 4,446 kg, as compared to a PbA system with equivalent functionality but higher weight (13,044 kg), due to the lower energy density provided by PbA batteries (Rydh and Sanden, 2005; Matheys et al., 2009).

System boundary - Case 2: For the PbA battery system, the life cycle stages considered were battery production, stationary application (i.e. charge discharge losses) and battery recycling. For the cascaded EV LIB stationary storage system, life cycle stages included were LIB production, battery refurbishment, secondary use and LIB recycling. If a utility is making a

direct comparison between a used LIB and a new PbA battery, then allocation is necessary to partition impacts of the initial LIB manufacturing (P1) and EOL management stages (E1 and E2) between the first and second (cascaded) lives since these activities are common to both the battery uses. Fig. 3.2 illustrates the analyzed life cycle stages of EV LIB with an extended use in stationary application, with a demarcation of stages for which environmental impact allocation is required.

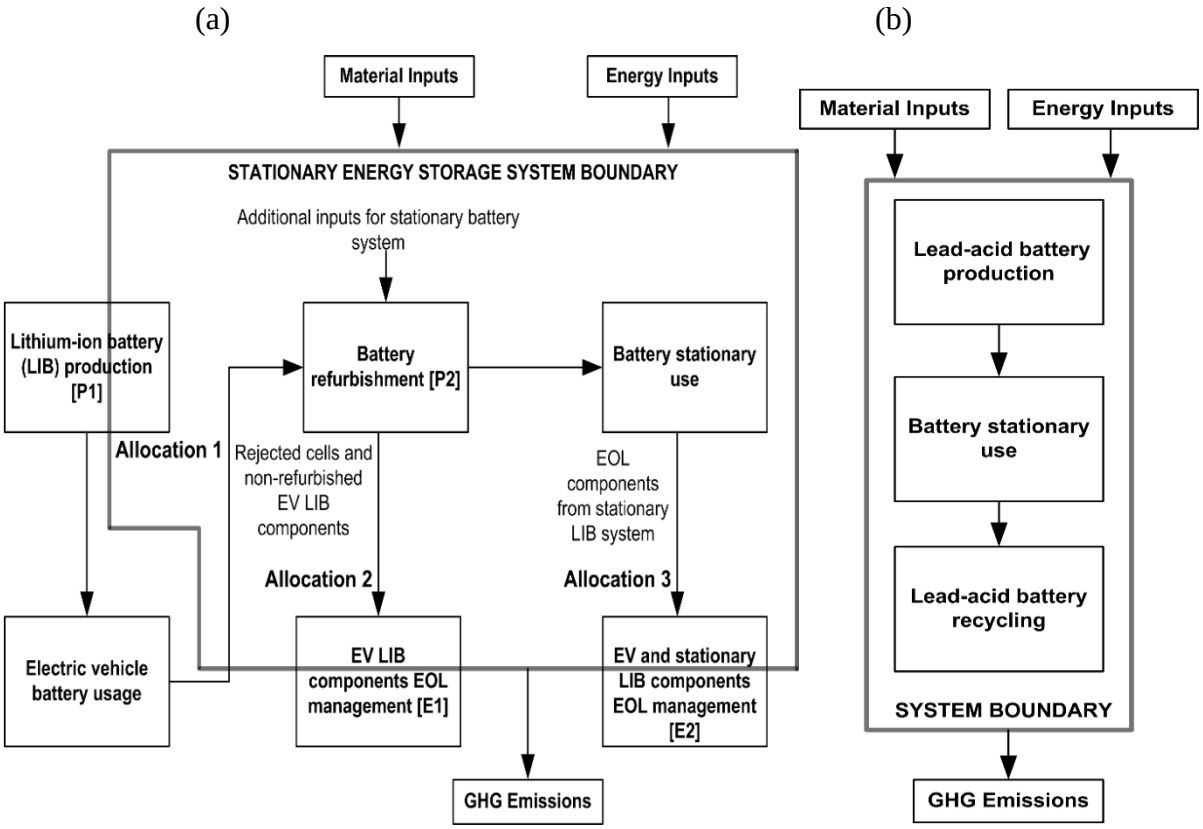


Figure 3.2 System boundary to compare environmental implications of battery choice for stationary energy storage (case 2). System (a) shows the allocation of upstream EV LIB production and subsequent EOL management impacts to the stationary energy storage system. System (b) shows the stages considered for the incumbent PbA system. These systems correspond to results shown in Figs. 3.5, 3.6, and 3.7

3.2.3 Life Cycle Inventory and Life Cycle Impact Assessment

All battery material production, processing and use phase stages were modeled in SimaPro LCA software using the ecoinvent database version 2.2 (ecoinvent Centre, 2010). U.S. based electricity grid mix was used for electricity input data. The environmental impacts considered were Cumulative Energy Demand (CED) and Global Warming Potential (GWP). For CED calculation, characterization factors and primary energy sources provided in SimaPro

software CED calculation methodology were used. For the estimation of GWP, all emissions contributing to GWP were considered and IPCC 100 year GWP impact factors were used. LCI data were obtained from a variety of sources including LCI databases, lab-scale tests, technical literature and previous LCA studies, as described below for each life cycle stage analyzed, with additional information provided in the Appendix B.

EV LIB production: The approach for modeling LIB production is process-based, similar to methodology employed by Dunn et al. (2012), U.S. EPA (2012) and Notter et al. (2010). This stage includes the manufacture of various EV LIB components such as the $LiMn_2O_4$ cathode, graphite anode, plastic separator, electrolyte salt (Lithium hexafluorophosphate [$LiPF_6$]), electrolyte solvent (Ethylene Carbonate), and battery management system (BMS) as well as processes involved in assembly of the cells and battery pack. (See also Section S.1 in the Appendix B).

EV battery use: The EV use phase of the LIB was modeled as the electricity lost due to battery efficiency over the lifetime of the EV and the additional energy needed to carry the weight of the battery, based on approaches adopted in previous studies (Van den Bossche *et al*., 2006; Zackrisson *et al*., 2010). At the beginning of life of EV LIB, a roundtrip efficiency of 95% has been assumed based on advanced vehicle tests for the Nissan Leaf battery (Garetson, 2013) which gradually reduces to 80% at battery EOL in EVs, which is defined as the point at which battery capacity fades to 80% (Ahmadi et al., 2014b). In addition to electricity consumption, the use phase also includes transport of the battery from the manufacturer to the car assembly site. (See also Section S.2 in the Appendix B)

EV LIB refurbishment: This stage includes the transport of EV LIBs to a refurbishment facility, testing of LIB cells to assess electrical performance for feasibility in stationary application, and repackaging cells into stationary battery modules (Cready et al., 2003). Apart from cells, some module components are assumed to be reused while additional components are added during refurbishment, such as copper connectors, battery cabinet and a new BMS (See also Section S.3 in Appendix B).

Stationary battery use: This stage includes transport of the battery (refurbished EV LIB or a new PbA battery) to the site of stationary usage as well as charge discharge electricity losses due to battery efficiency during operation. Capacity fade with aging and the corresponding energy efficiency fade for the refurbished EV LIB based energy storage system has been modeled similar to Ahmadi et al. (2014b). For the PbA battery, a direct correlation between increase in battery internal resistance and increase in efficiency losses with aging is considered (See also Section S.4 in Appendix B).

LIB EOL management: The recycling of cells and module components from an EV battery pack occurs at two stages. First, recycling may be required for any rejected cells coming out of EV use that are deemed unsuitable for stationary application. At the same time, many EV LIB pack components such as the battery jacket and BMS would be separated and recycled during refurbishment. Second, once batteries reach their final EOL after stationary use, all cells and additional components added to the stationary battery during refurbishment (e.g. BMS, copper connectors) would be recycled. Non-recyclable materials such as mixed plastics, graphite, electrolyte, binders etc. are considered to be sent to the landfill as indicated in Chapter 2 (Richa et al., 2014). Recycling impacts and recovery is modeled assuming a mix of hydrometallurgical and pyrometallurgical processing (Mudgal et al., 2011), with credit provided for avoiding the production of primary material based on calculated recovery potential. (See S.5 in Appendix B)

PbA battery production: The material composition of the PbA battery was estimated from Rydh (1999), Rantik (1999) and Sullivan and Gaines (2012). This included lead inputs (for active material, grid, and poles), electrolyte, plastic cases and covers, glass separator and copper connectors. A 47:53 mix of recycled and virgin lead was employed (Hittman Associates, 1980; Sullivan and Gaines, 2012). Energy inputs for PbA battery manufacturing was obtained from Hittman Associates (1980). (See also Section S.6 in the Appendix B).

PbA battery recycling: A smelter based process for secondary lead production was modeled (ecoinvent Centre, 2010; Fisher et al., 2006) and recycling credit was provided for avoiding the production of virgin lead, according to the initial fraction of lead from primary and secondary sources specified for PbA battery production.

3.2.4 Sensitivity Analysis

The LCA model is sensitive to choices made when defining model assumptions and specifying technical parameters in battery reuse. These parameters are characterized with a high-degree of uncertainty, as the feasibility of reuse is still an emerging area of research and application. To capture likely areas of uncertainty, key technical parameters are explored further, which are selected on potential performance constraints of second use LIBs (Cready et al., 2003), variability in PbA battery types available in the market, and data availability. The three variables meeting these criteria are described below. Other parameters, like degradation of LIB efficiency over time and in different applications, are clearly important factors in comparison to new PbA cells, but are outside the scope of this study, as technical studies have not yet generated sufficient data to fully characterize sensitivity in LCA.

Cell conversion rate (F) represents the percentage of EV LIB cells that can feasibly be repurposed for stationary application. The failure to convert cells between life cycles, (thus leading to EV LIB cells being "rejected" for stationary use) may be due to either conditions during the EV life cycle (temperature, driving patterns, charging frequency) or to specific conditions that must be met (capacity) for use in stationary energy storage systems (Cready et al., 2003; Neubauer et al., 2012). To reflect the importance of the parameter F and to capture the high degree of uncertainty around its value under different operating conditions, a wide range of values (between 10-100%) for F was analyzed. The baseline model considered a mid-range scenario of 50% F value (Table 3.1), which falls between the extremes of either the entire battery pack being rejected or being viable.

In the extended life cycle case, F would determine the size of the stationary battery system resulting from a single EV battery. For the stationary energy storage case, F would determine the number of EV LIB cells required to build a single stationary battery with equivalent functionality to the PbA alternative.

Refurbished LIB lifespan represents the calendar years after which refurbished EV LIB cells fail to meet specific performance criteria in stationary application. Typically, how often the battery is charged would depend on the stationary application and the demand for battery stored

energy (Cready et al., 2003). Similar to previous studies (Ahmadi et al., 2014a; Ahmadi et al., 2014b; Rydh and Sanden, 2005), a single daily charge-discharge cycle over the battery lifespan has been assumed to demonstrate a generic example of battery operation. Recent studies have assumed a lifespan of refurbished EV LIBs in stationary application ranging from 1 to 10 years (Ahmadi et al, 2014a; Cicconi et al., 2012; Beer et al., 2012; Cready et al., 2003; Narula et al., 2011). Hence, this parameter has been varied across the same scale, resulting in a cycle life of 365 (1 year lifespan) to 3650 (10 year lifespan) charge-discharge cycles. This range should capture the wide variability expected due to vehicle use patterns and climate extremes during the automobile use phase of the LIB. The lifespan of PbA battery is considered to be 5 years in all scenarios based on the DoD range (Rydh and Sanden, 2005; Bindner et al., 2005). In the extended life cycle case, the lifespan determines the total number of refurbished LIB based systems that can replace a corresponding number of equivalent functionality 5 year lifespan PbA batteries. For the stationary energy storage system case, the lifespan parameter determines the number of system replacements needed to provide the 20 year use specified by the functional unit.

PbA battery efficiency is usually between 70% and 85% (Rydh and Sanden, 2005; Matheys et al., 2009; Matheys and Autenboer, 2005; Albright et al., 2012; Parker, 2001; Celik et al., 2008; Rydh, 1999), depending on battery type and manufacturer. This range determines the low and high efficiency of PbA batteries with a mid-range baseline efficiency of 77.5%. This parameter determines the relative energy used or lost during use of the stationary energy system. Similar to LIBs, the PbA battery efficiency is also modeled to decline gradually with aging as explained in the Appendix B (Section S4).

Apart from these parametric variations, the comparison among LCA allocation methods provides for an assessment of sensitivity to methodological choices (specific to Case 2) which is described in the next section. The parameters used for sensitivity analysis based on these criteria, indicating best to worst scenarios for EV LIB reuse are provided in Table 3.1.

Table 3.1: Parameters varied for sensitivity analysis

Sensitivity criteria	Values/Methods	Best Case Scenario	Base Case Scenario	Worst Case Scenario	Case applicable to
Cell conversion rate	10% to 100%	100%	50%	10%	Extended life cycle (Case 1), Stationary energy storage (Case 2)
Refurbished LIB lifespan	1 to 10 years	10 years	5 years	1 year	Extended life cycle, Stationary energy storage
PbA battery efficiency	70% to 85%	70%	77.5%	85%	Stationary energy storage [a]
Allocation	50/50, Quality-based; Cut-off	Cut-off	Cut-off	50/50	Stationary energy storage

[a] For the extended life cycle (case 1), the baseline PbA efficiency (77.5 %) is applied in all scenarios

3.2.5 Allocation

The cascading reuse of refurbished EV battery has been considered similar to open-loop recycling, which is a special case of a multi-functional system (ISO, 2006). In open-loop recycling, the material under consideration is recovered for use in a different product than the one from which it is recycled. Similarly, in the case of cascading reuse, the product (the EV battery) is reused in an application that has a different function and use than the first life cycle. Here, the need for allocation specific to an "open-loop product cascade" – a concept not yet fully explored in LCA – is based on existing open-loop recycling allocation procedures (quality based, cut-off, and 50/50; Ekvall and Tillman, 1997), but with case-specific modifications. For allocating impacts of EV LIB production and EOL management to the stationary battery, only those components that were considered for reuse (i.e. EV LIB cells and module components) were accounted for and life cycle impacts of any components rejected or recycled during LIB refurbishment (BMS, battery jacket) were not allocated to the stationary battery. In this section, the following nomenclature is applied:

- I_{bs} = Environmental impacts of EV LIB cell and module production and EOL management that are allocated to stationary energy storage system
- $I_{e,v1}$ = Environmental impacts of EOL management of EV LIB cells and module components not used in stationary battery (*v1*)
- $I_{e,v2}$ = Environmental impacts of EOL management of EV LIB cells and module components used in stationary battery (*v2*)

- $I_{pn,v}$ = Environmental impacts of producing all EV LIB cells and module components, (v = $v1+v2$)

The overall environmental burden of the refurbished EV LIB based stationary energy storage system would include the allocated environmental impacts due to EV LIB cells and modules production and EOL management (I_{bs}), EV battery refurbishment, efficiency losses due to use phase and EOL management of additional stationary battery components added during refurbishment. Different allocation approaches for determining I_{bs} are explained:

a) Quality based allocation: This approach is usually based on physical qualities such as material mass, purity, and energy content or economic attributes of the virgin and recycled material in the case of open-loop recycling (Knoepfel, 1994; Ekvall and Tillman, 1997; Kim et al., 1997; Nicholson et al., 2009; Borg et al., 2001; Guinee et al., 2004, etc). In the case of EV LIB reuse, the cogent properties are associated with the battery's chemical and physical properties (e.g., energy storage capacity), which determine the extent to which the used LIB can meet the functional demands of secondary energy storage. The market price in the two life cycles also reflects the value of refurbished stationary LIB relative to the EV LIBs. The environmental burden for production of EV LIB pack components ($I_{pn,v}$) that are considered for reuse (i.e. cells and module components) is clearly shared to some degree by both battery systems. While the impacts of some EOL processes ($I_{e,v1}$) occur prior to the construction of the stationary battery, they are the result of some EV LIB cells and module components being rejected for stationary use, and thus the stationary system bears some part of this burden (or incentive in the case of recycling credits). It is possible that the utility sector would be responsible for some part of this EOL management, especially if the infeasibility of the cells for stationary use is detected towards the final stages of testing and not in a preliminary analysis in a battery collection center. The second aspect of LIB EOL is associated with the waste management of EV LIB components used in both the initial and cascaded life cycles. Hence, the environmental impacts associated with these life cycle stages, $I_{e,v2}$ would be shared between the two battery systems. Based on these arguments, the two quality based allocation approaches are described:

Energy storage based allocation: As LIB cells are passed on from their first to second life, there is also an "export" of energy storage capacity from the more demanding EV application to the

stationary energy storage application. Using energy-based allocation to partition environmental impacts between the two battery systems provides a reasonable basis for allocation since it gives a clear indication of this "function transfer." The energy storage of the stationary battery (E_s) is 450 kWh based on the functional unit for case 2. The energy storage (E_{ev}) of the EV battery system was determined by the number of EV LIBs required to build a single stationary battery, accounting for any fraction of LIB cells that were rejected for reuse. The factor ($E_s/(E_s+E_{ev})$) represents the energy storage quality of the stationary battery system with respect to the overall quality of the multifunctional system. Based on this factor and the quality based allocation methodology suggested by Ekvall and Tillman (1997), environmental impact, I_{bs}, allocated to the stationary battery due to EV LIB cells and modules production and EOL management would be:

$$I_{bs} = \frac{E_s}{E_s + E_{ev}} \left(I_{pn,\mathrm{v}} + I_{e,v1} + I_{e,v2} \right) \qquad (1)$$

Market price based allocation: The use of market price for new and refurbished LIBs in terms of \$/kWh combines considerations of the energy storage function and the market price of the battery. This allocation is useful for both stakeholder groups, since it reflects the functional quality of the LIB within a techno-economic system comprising the EV and utility sectors. The price of a retired EV LIB would not be reduced by the same percentage as its capacity fade post-EV application. For this study, the selling price of refurbished EV LIB was considered to be 30% of future new EV LIB cost in terms of \$/kWh based on Neubauer et al. (2012).

V_{ev} is defined as the initial value of the EV battery system comprising of multiple LIB packs and is calculated as the product of energy storage capacity of EV battery system (i.e. E_{ev}) and the price in \$/kWh for a new EV LIB pack. V_s is the market value of the stationary battery and is defined as the product of energy storage capacity of stationary energy storage system (E_s) and the price in \$/kWh for a refurbished EV LIB. The factor V_s/V_{ev} represents the market value of the stationary battery system with respect to the market value of multiple LIBs that constituted the EV battery system. Similar LCA allocation methodology based on economic value or quality reduction parameters has been suggested in prior studies (Werner et al., 2007; Borg et al., 2001; Karlsson, 1994, etc.). In this approach, I_{bs} would be:

$$I_{bs} = \frac{V_s}{V_{ev}} \left(I_{pn,\mathrm{v}} + I_{e,v1} + I_{e,v2} \right) \qquad (2)$$

b) Cut-off allocation: Cut-off allocation methodology is based on the argument that each product should only be assigned the environmental impacts directly caused by its life cycle (Ekvall and Tillman, 1997; Werner et al., 2007). In this case, cut off implies that an EV LIB was manufactured primarily for vehicle application and no impacts of initial battery manufacturing or EOL management not leading to reuse are allocated to the stationary energy storage system. Since the second level of LIB EOL management processes ($I_{v2,\ e}$) occur at the EOL of the stationary battery, for which the stakeholder in the utility sector are likely to be responsible, their burden should be borne completely by the stationary system. Cut-off represents the best case scenario for the stationary energy storage system, as the raw materials are, in effect, "free" of impact. Based on the above arguments, the following allocation method expresses the environmental impacts, I_{bs}:

$$I_{bs} = I_{e,v2} \qquad (3)$$

c) 50/50 allocation: This allocation scenario was selected to represent a conceptual "worst case" for allocation to the stationary energy system, implying that the LIB manufacturing and EOL impacts assigned to the cascaded system would not be any higher than those assigned to the original application for which the battery was designed and produced. The underlying argument is that EV application and stationary application are both part of the same life cycle of LIB, which could become increasingly valid if reuse partnerships between EV and utility are arranged in advance, hence all environment burden of LIB production and EOL management would be shared equally between the EV battery system and the stationary battery system:

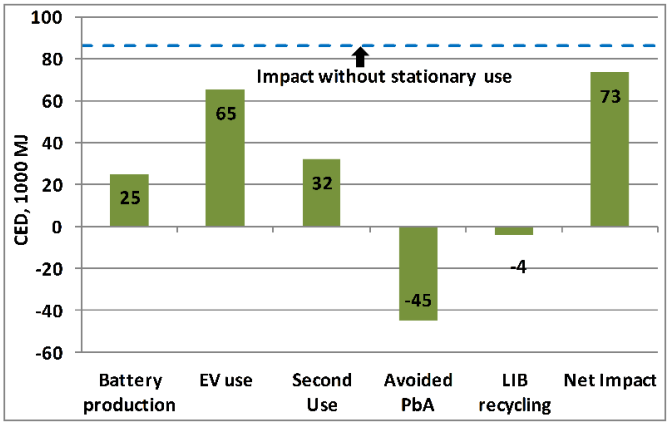
$$I_{bs} = 0.50 * \left(I_{pn,\mathrm{v}} + I_{e,v1} + I_{e,v2}\right) \qquad (4)$$

3.3 Results and Discussion

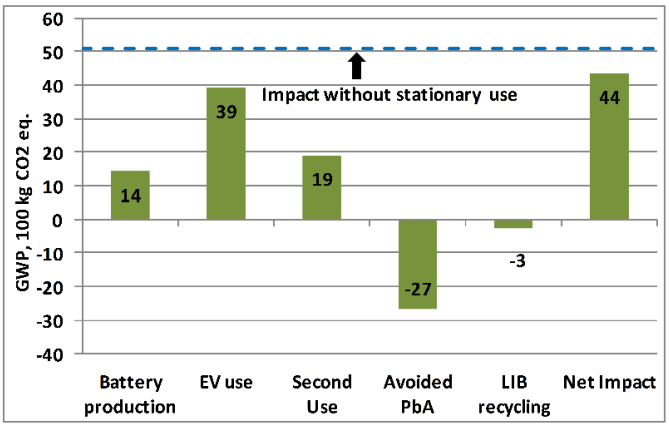
3.3.1 Extended life cycle (Case 1): Results show that EV use phase followed by LIB production are the life cycle stages contributing to the highest impact for the LIB (Fig. 3.3). This trend is consistent with previous LCA studies that have estimated GHG emission impacts of EV LIBs (Zackrisson et al., 2010; Samaras and Meisterling, 2008; Hawkins et al., 2013). The CED and GWP of EV battery production was calculated at 1037 MJ/kWh and 59 kg CO2eq./kWh, respectively. These findings are close to the estimations made by other process-based LCA studies which range from 613-948 MJ/kWh and 50-62 kg CO2eq./kWh for CED and GWP

impacts (Notter et al., 2010; U.S. EPA, 2012). Variability in EV use phase impacts are observed due to differences in the choice of electricity grid mix, modeled parameters such as number of cycles during battery lifespan or the distance travelled by the EV during its lifetime (Ellingsen et al., 2014), and system specifications which include considering total energy used for charging the LIBs (Samaras and Meisterling, 2008; Hawkins et al., 2013) or accounting for only the energy used to carry the battery weight and charge discharge losses (Zackrisson et al., 2010).

Extending the battery's life by stationary use introduces trade-off between the environmental cost of LIB refurbishment and use and the benefit of avoiding the production and use of a stationary battery based on existing PbA technology. For the baseline scenario, which assumes a 50% cell conversion rate from EV to stationary application and a 5-year refurbished LIB lifespan, the environmental benefits outweigh potential costs, providing an overall CED and GWP credit of 12,850 MJ and 740 kg CO2-eq, respectively; a net reduction of15% in these impacts over the EV LIB life cycle (Fig. 3.3).



(a)



(b)

Figure 3.3 CED (a) and GWP (b) impacts of an EV LIB with cascading reuse. These results reflect the baseline assumptions that 50 % of the EV LIB cells can be converted for stationary use application and that the stationary

battery will have a 5-year life span. For comparison purposes, the shading used above is applied to denote the "baseline" results in subsequent figures. The dotted line reflects the impact of the EV LIB life cycle with no reuse in stationary application

Sensitivity analysis on parameters applicable to Case 1 (see Table 3.1), demonstrated that net environmental credits due to EV LIB reuse reduced with decreases to both cell conversion rate (F) and refurbished LIB lifespan. This result is due to the environmental burden of transporting and testing more cells than required by the stationary energy storage system and multiple replacements of cells in the stationary battery. However, overall reduction in EV LIB environmental impact was observed for almost all scenarios (Fig. 3.4), with reductions from as low as 0.3% to as high as 69% for CED, across the range of all scenarios. Similar reductions (between 0.2 to 68%) were observed in GWP (Fig. S1 in Appendix B). For a very few extreme scenarios (notably 1-year lifespan and cell conversion rates less than 50%), a small net increase (<3%) in these environmental impact categories was shown (Fig. 3.4). These results suggest that second use of EV LIBs is capable of further amplifying the environmental benefits accrued due to adoption of EV technology, which more broadly can enable CO_2 emission reductions of about 20-50% when compared to conventional gasoline vehicles (considering country specific/regional grid mixes for battery charging) (Samaras and Meisterling, 2008; Sustainable Energy Ireland, 2007; Hawkins et al., 2013). Reductions in environmental impacts are expected to parallel decreases in battery cost adjacent to technological progress. The U.S. Department of Energy has estimated that by 2020, used EV batteries would be available in significantly large quantities to reduce LIB costs by 70% and meet targets as low as $125/kWh (Howell, 2012; Neubauer and Pesaran, 2011).

EV LIB recycling is likely to provide minor environmental benefit: 8-9% of total environmental credits over the extended life cycle of the battery in the baseline scenario (Fig. 3.3). One barrier to EV battery recycling routes has been the gradual adoption of low cost LIB chemistries (e.g. manganese oxide and iron-phosphate) by EV manufacturers, which result in lower material recovery value than cobalt based chemistries (Wang et al., 2014a, Wang et al., 2014b). Declining economic motivation for recycling, in parallel to the growing trend in battery landfill bans in the U.S. and Europe (CA Code, 2006; New York State Rechargeable Battery Law, 2010, European Commission, 2006), and extended producer responsibility for vehicle EOL in Europe (EU, 2000) all underscore the importance of second use applications for EV LIBs.

Many OEMs are actively considering proactive EOL management options, including stationary storage, in advance of widespread EV deployment (Doggett, 2011; Williams, 2011; Kuo, 2011).

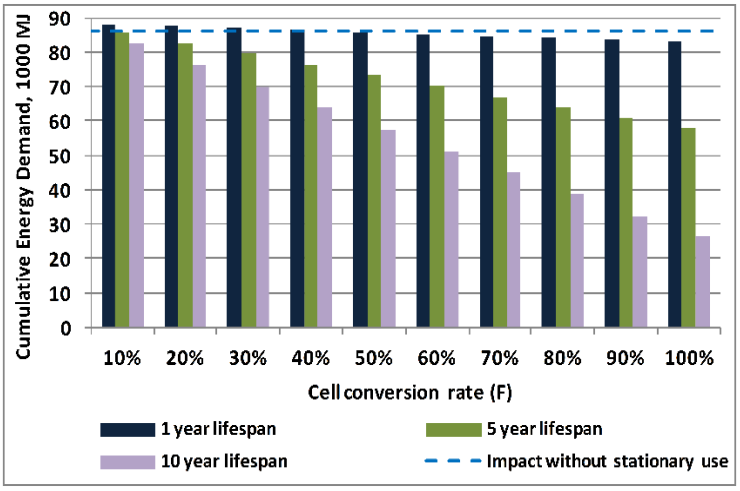


Figure 3.4 Sensitivity analysis: CED of EV LIB with extended life under different scenarios of cell conversion rate and refurbished EV LIB lifespan

3.3.2 Stationary Energy Storage (Case 2): In a direct comparison of reused LIB to new PbA battery systems for stationary energy storage, the baseline scenario indicates that CED and GWP impacts of refurbishing EV LIBs for a five year service life in stationary application (i.e. 4 battery replacements during the total system lifetime) represent less than half of the impacts of manufacturing a new PbA battery. Particularly, the use phase GHG and CED impacts of the refurbished LIB system were 21% lower than the PbA battery with mid-range (baseline) energy efficiency of 77.5% (Fig. 3.5). However, if the incumbent battery system had a significantly higher beginning-of-life efficiency (85%), these use-phase impacts of the LIB system were actually higher by about 8%. Recycling the PbA battery provides no environmental credit, as the lead used to produce these systems already has a high proportion of secondary content. On the other hand, for LIB recycling, there is some environmental credit obtained due to metals recovery, assuming closed-loop recycling. In general, the baseline scenario refurbished EV LIBs were found to be environmentally favorable than PbA systems, even including the most efficient technology for the latter. A net reduction of 13 to 46% in CED and 12 to 46% in GWP impacts was estimated for the stationary LIB in the baseline scenario relative to equivalent functionality PbA battery over a range of energy efficiency scenarios for the latter (Fig. 3.5; For GWP impacts refer to Fig. S2 in section S7 of Appendix B).

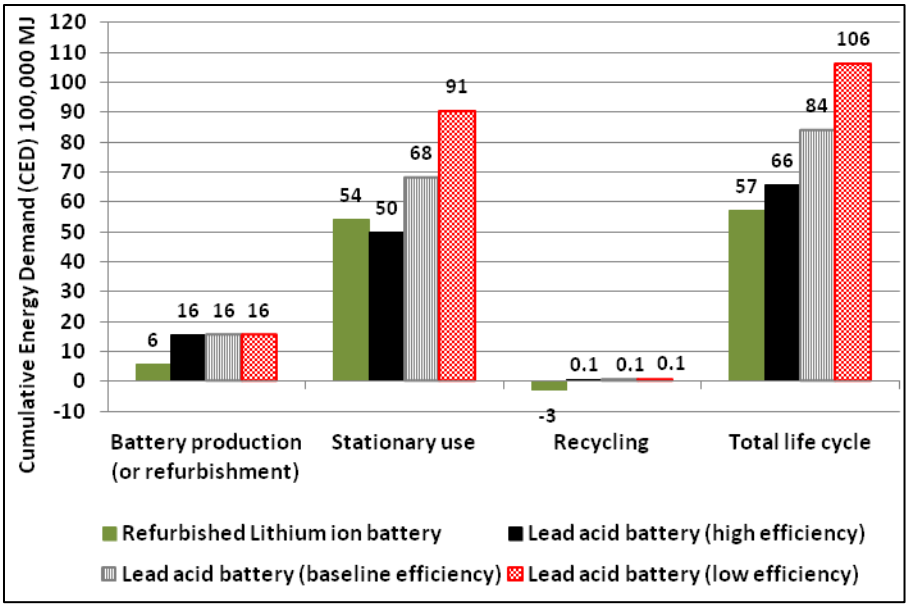


Figure 3.5 CED of stationary energy storage system, comparing the baseline LIB scenario to three scenarios of PbA battery efficiency

These environmental benefits could potentially be reduced under scenarios in which the refurbished LIB does not provide the reliability required by stationary energy storage or if a high volume of cells is rejected during refurbishment (Fig. 3.6). The best outcome for the refurbished stationary battery (using cut-off allocation) was that of 100% F and 10 year lifespan of refurbished cells, requiring only a single replacement during the lifespan of the stationary battery system. For these parameters, a 49% reduction in the CED impact of the stationary battery is possible when compared to low efficiency PbA batteries in the market (Fig. 3.6(a)). It is likely that these LIBs with high reuse potential could be obtained from EV applications where the vehicle itself reaches EOL before the battery (e.g., in cases of accident) or the battery is replaced early due to EV resale (Richa et al., 2014).

A prominent factor that can define reuse feasibility and environmental performance is the allowable DoD of second use LIBs, which would govern sizing of these stationary energy storage systems relative to equivalent functionality PbA batteries. In this study, the DoD of both the stationary PbA and LIBs were assumed to lie in the range of 33-42% and hence both systems are considered to be of equal size (i.e. 450 kWh), which is extremely conservative for LIBs. In general, new LIBs can provide deep discharges of as high as 80% (Rydh and Sanden, 2005; Albright et al., 2012). However, there is immense uncertainty over the safe DoD range for second use LIBs, with previous studies assuming DoD within a broad range of 20-80% (Ahmadi

et al., 2014a; Neubauer et al., 2012; Neubauer and Pesaran, 2011; Ahmadi et al., 2014b). Robust and more stable LIB chemistries, such as lithium iron phosphate ($LiFePO_4$), (Xu et al., 2010; Choi et al., 2010; Krieger et al., 2013) that have demonstrated slower capacity fade compared to manganese spinel based chemistry (Han et al., 2014) are likely to be operated at higher DoD (Ahmadi et al., 2014b). However, a moderate DoD range for Mn-based batteries is a reasonable assumption before performance uncertainties of second use LIBs can be resolved. If these LIBs can be operated over a higher DoD range (65-80%) in stationary application, then a much smaller sized LIB system would be needed (approximately 230 kWh to deliver 150kWh of energy on a daily basis). In that instance, the CED impact for the stationary energy storage system (Case 2) can be reduced by as much as 74% (instead of 49% for the 450 kWh LIB) when compared to a low efficiency 450 kWh PbA battery system.

Second-life batteries are currently ineligible for incentive programs or federal investment tax credits for grid storage, onsite or residential energy storage systems in the U.S. However, quantifying the environmental benefits along with economic gains can stimulate policy support to promote cascaded battery installations to help meet the ambitious renewable energy generation and energy storage targets in U.S. states like California (Elkind, 2014). The U.S. DOE has already made investments in projects to test the feasibility of such systems (Neubauer and Pesaran, 2011). Similarly, partnerships have been undertaken by automakers such as Nissan, General Motors (GM) and BMW and utility providers such as ABB, Sumitomo and Vattenfall towards collaborative research efforts (Bond, 2013; Doggett, 2011; Sumitomo Corp., 2011; Gordon-Bloomfield, 2015). In fact, the scenario of 100% cell conversion rate and long lifespan can be considered similar to business models in which entire LIB packs are reused and where significant information is available about battery performance during EV use (e.g., as captured by systems like OnStar for the GM Volt). One example is the collaborative pilot project between ABB and GM resulting in a prototype backup power storage unit from five used GM Volt batteries that can power 3-5 homes for 2 hours (Bond, 2013).

The worst environmental performance of the refurbished stationary LIB estimated under the cut-off allocation methodology was that of 10% F and 1 year refurbished LIB cells' lifespan. This scenario saw a three-fold increase in the environmental impact of the stationary energy storage system when compared to high-efficiency PbA battery system. Such low performance LIB cells might result from highly cycled EV batteries, cells used in temperature extremes, or

those of the highest age on a potential lifespan distribution as demonstrated in Chapter 2(Richa et al, 2014). However not all scenarios of low cell conversion rate offset the environmental benefits of EV LIB reuse (Fig. 3.6 (a)). A sufficient lifespan in stationary use is capable of compensating for some of the environmental losses incurred due to a large number of EV LIB cells found to be technically infeasible for stationary application. For example, even with an extremely low F of 10%, the stationary battery based on refurbished cells was estimated to have a life cycle CED comparable to a high efficiency PbA battery if a 10 year lifespan of refurbished cells was attained. For extremely low lifespan of refurbished cells (1 year), the cut-off for environmental feasibility was found at F values of higher than 50%, when comparing to a high-efficiency PbA battery. However, this criterion can be relaxed when comparing to low-efficiency PbA batteries. Conversely, high F value can enable second use LIBs with poor lifespan to be environmentally superior to PbA based systems. In terms of real world applications, cascading whole EV LIB packs, even with extremely low lifespan, would still be environmentally preferable for less intensive stationary storage or ad-hoc applications. These can include low demand (~twice per year) power back up systems such as those for telecommunication applications.

However, environmental feasibility is not the only criteria that would determine the adoption of retired EV LIBs in utility based applications. Low conversion or low lifespan of used EV LIB cells is likely to be accompanied with high refurbishment, battery maintenance and replacement costs. In general, a PbA battery system cost lies in the range of 65-120 $/kWh depending on the technology (Albright et al, 2012). The pricing of second use stationary LIB is uncertain, but may range from 38 to 132 $/kWh, depending on future EV adoption and new LIB costs (Neubauer et al, 2012). Additionally battery re-installation costs can vary from 4-17 $/kWh with labor cost being as high as 100 $/hour to replace a LEAF sized battery (Neubauer et al, 2012). These factors would ultimately influence the economic feasibility of low lifespan retired EV LIBs when compared to PbA battery based systems, and need to be further analyzed to estimate the overall eco-efficiency of LIB reuse.

Additionally, this study presents a relatively conservative estimate for EV battery refurbishment by analyzing a system in which refurbishment would require disassembly and testing of the individual cells contained in EV battery packs (Nenadic et al. 2014). However, a more realistic approach will likely be based on testing battery modules (or even whole packs) instead of cells which could result in lower electricity inputs during LIB refurbishment.

However, module level testing can be accompanied with additional environmental trade-offs as the energy used during refurbishment may decrease by as much as 27-41% (Cready et al. 2003), but may result in aggregate diagnostics that result in a lower "module yield rate" (Neubauer et al., 2012), leading to a large number of technically viable cells entering the waste stream.

3.3.3 Sensitivity to Allocation Choices

Beyond sensitivity to technical parameters, tradeoffs between the two battery systems compared also depends on the method of allocating upstream impacts for the EV LIB production and EOL management. While cut-off allocation, shown in results of Fig. 3.6(a), is the best scenario for refurbished LIB based stationary energy storage system, a choice of 50/50 allocation results in a higher impact for the secondary life (Fig. 3.6(b)). In many instances, the cut-off points beyond which a refurbished battery was no longer the preferable option were changed substantially. For example, now for a 5-year lifespan refurbished LIB, a minimum F of 20% or 95% conversion rate of EV LIB cells would be required to be environmentally comparable with a low or high efficiency PbA battery, respectively. For cut-off allocation, these conversion rates were as low as 5-19%.

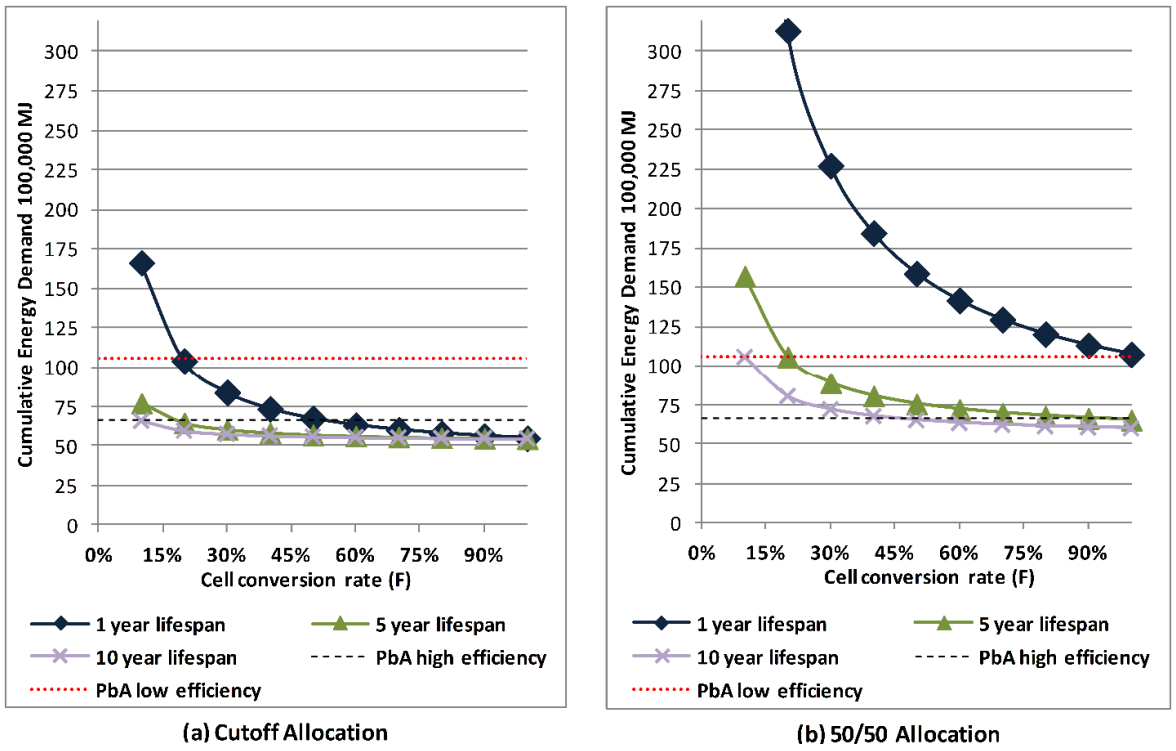


Figure 3.6 Impact of cell conversion rate and refurbished LIB life span on environmental feasibility of EV LIB reuse (CED impacts) under (a) cutoff allocation scenario and (b) 50/50 allocation scenario

When comparing all allocation methods tested, quality-based approaches resulted in intermediate environmental performance. For example, for the baseline scenario refurbished LIB, allocating based on market price and energy storage resulted in net CED impacts that were 12-21% higher, respectively, than calculated using cut-off, but 16-9% lower, respectively, than 50/50 (Fig. 3.7). The difference in the environmental impact of stationary battery under different impact allocation methods diverges further under scenarios of extremely low cell conversion rate. In general, allocating based on market prices of the initial and reused LIBs resulted in lower impacts assigned to the stationary energy system than when using energy storage as the quality indicator. However, it must be noted that any forecasts of prices are estimates only, as a reuse market has not yet been established at this early stage of EV adoption. For the utility sector, the choice of allocation method could primarily depend on the parameter (i.e. energy or price) that determines their perceived value of a cascaded LIB battery. Allocation could also reflect by the battery ownership and EOL liability in reuse cascade. Since neither the EV market nor the battery technologies are fully evolved yet, it may take some time for an Extended Producer Responsibility (EPR) network to be established in this domain. In general, the EPR principle is an integral part of EU waste directives such as WEEE, Battery and ELV directives (European Commission, 2014) – regulations which can potentially cover EV LIBs in future. The recycling of spent LIBs would involve different actors from the EV sector, namely the battery material producers, battery producers, vehicle producers, vehicle dealers, and vehicle treatment operators (Hoyer et al, 2011). However, once the reuse application becomes business as usual, new actors from the utility sector will be involved as well. Modeling choices must evolve to reflect changing stakeholders in this dynamic system.

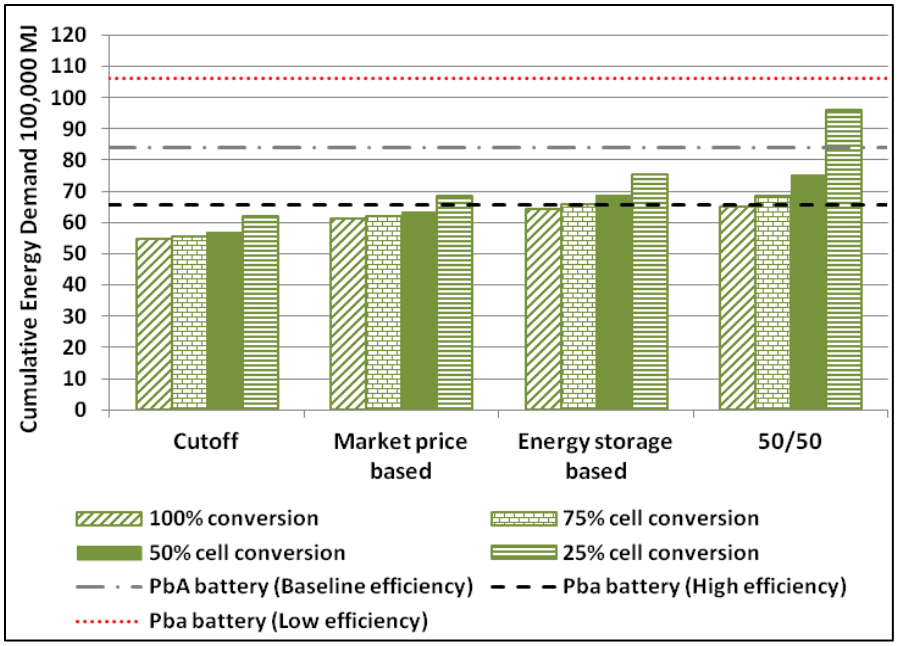


Figure 3.7 CED of refurbished EV LIB-based energy storage system under different allocation approaches (for a 5-year battery life span in stationary application)

3.4 Conclusions

Use of refurbished EV LIB cells in energy storage applications shows promise of reducing the environmental impacts of these systems. Such reductions would be primarily due to avoiding the life cycle of an equivalent PbA battery based system. Additionally, lifespan extension would delay the entry of EOL LIBs into the waste stream, creating ancillary benefits by allowing additional time for capacity building in the nascent LIB recycling sector. A wide array of technical and modeling choices were analyzed here, and despite variability in results, LIB reuse almost universally creates benefit either to the EV sector by extending battery life and improving the competitiveness of EV technology or to the stationary energy sector by avoiding production and use of a less-efficient PbA system. However, given the prospective nature of this study, there is still immense uncertainty, particularly related to how closely actual battery reuse parameters will align with modeling choices applied here.

Additional improvements can be gained via engagement and partnerships by both stakeholder groups. One promising approach may be to directly engage vehicle battery OEMs in rebuilding the packs for the secondary application. This approach leverages historical knowledge of the cells such as their usage and deep discharge event, which has the potential to minimize or even obviate the expensive process of refurbishing. Furthermore, the engagement of OEMs in

grid applications may alleviate apprehensions associated with usage of vehicle batteries as an additional storage for smart grid applications. Battery OEMs and refurbishers may adopt different strategies depending on LIB pack design, economic viability, or policy incentives.

Comparisons of cascaded use of retired EV LIBs with PbA battery technology were made since currently the latter is the most widely used energy storage technology for grid support, primarily owing to its low cost (Albright et al., 2012; Soloveichik, 2011). However, it is expected that both redox-flow batteries and high temperature sodium batteries are going to be widely adopted by the utility sector in future because of their higher energy density, electrical efficiency and lower maintenance which can make them cost competitive for stationary applications (Dunn et al., 2011; Doughty et al., 2010). Furthermore, as the market for lithium-ion batteries grows both for consumer electronics as well as for electric vehicles, they could be a feasible option for grid based energy storage applications due to the economies of scale expected. Hence, further research is required to compare the environmental and economic benefits of cascaded use of retired EV LIBs with respect to new LIBs and other upcoming battery technologies.

Many uncertainties still exist, and empirical and modeling studies are necessary to further refine the estimates surrounding cascaded battery reliability, incumbent technology displacement, and scale up of battery reuse and recycling sectors. As this information becomes available, the LCA model and uncertainty analysis shown here can be iteratively improved. However, this proactive analysis of battery reuse alternatives allows stakeholders in both the EV and utility sectors to plan more effectively for the design, technology requirements, and business models needed to enable economically and environmentally feasible reuse strategies. More broadly, the methodological contributions to modeling allocation in cascaded product systems can also be applied in other sectors where components or whole products are repurposed and applied in a new application (for e.g. refurbished solar modules used in smaller scale applications) or even “upcycled” into value added product life cycles.

CHAPTER 4: ECO-EFFICIENCY ANALYSIS OF A LITHIUM-ION BATTERY WASTE HIERARCHY INSPIRED BY CIRCULAR ECONOMY

4.1 Introduction

While use of electric vehicles (EVs) can reduce dependence on fossil based transportation fuels and may ultimately curb carbon dioxide emissions, a major concern is the management of potential wastes generated when the lithium-ion batteries (LIBs) from EVs reach their end-of-life (EOL) (Richa et al., 2014; Wang et al., 2014b). The material flow analysis (MFA) of EOL EV batteries in Chapter 2 predicted a waste stream of 4 million metric ton (mT) of LIB cells between years 2015 and 2040 (Richa et al., 2014). Considering the pronounced scale of LIB usage as the number of EVs increases, a well-defined, proactive EOL management strategy is needed for these batteries. Such a strategy can be informed by "circular economy" principles such as reuse, recycling and landfill disposal (Ramoni and Zhang, 2013). A circular or closed-loop economy aims to eliminate waste by cycling materials and products within the system to achieve resource and energy efficiency as well as profitability (Ellen MacArthur Foundation, 2013; McKinsey & Company, 2014; Allwood et al., 2012; Allwood et al., 2011; Gregson et al., 2015; Ghisellini et al., 2016). Hence, both reuse and recycling propagate this concept by enabling a resilient infrastructure of LIB materials by avoiding primary metal production and reducing landfill disposal of potentially hazardous materials.

While few companies (e.g. Retriev, Chemetall, Umicore and Recupyl) commercially recycle LIBs, a significant body of knowledge has explored novel technologies for recovering constituent metals at bench scale including both hydrometallurgical and pyrometallurgical recycling pathways (Espinosa et al. , 2004; Xu et al., 2008; Dunn et al., 2012; Georgi-Mascheler et al. 2012; Ramoni and Zhang, 2013; Hendrickson et al., 2015; Nan et al. 2005; Dorella and Mansur 2007; Swain et al. 2007; Ferreira et al. 2009; Li et al. 2009; Chen et al. 2011; Li et al., 2013, etc.). While life cycle assessment (LCA) studies confirm the environmental benefit from recovery of LIB materials along different recycling pathways (Dewulf et al., 2010; Hendrickson et al., 2015; Dunn et al., 2012, 2015; Amarakoon et al., 2013), recycling will never be 100% efficient and it can only recoup a fraction of the embodied energy for materials – it does not even address energy input to battery assembly and manufacturing steps. Though LIB recycling could serve as an enormous source of high value materials as evident from the results in Chapter 2 (Richa et al., 2014), major economic barriers to commercial EV LIB recycling may emerge,

particularly potential transitions away from cobalt-rich battery chemistries (where cobalt drives the economic revenue of recycling) to lower cost chemistries (e.g. manganese oxide and iron-phosphate) by EV manufacturers (Wang et al., 2014b; Richa et al., 2014) and energy intensive pyrometallurgical recovery processes (Fisher et al., 2006).

Despite the wide commercial and research attention on recycling, it may not be the first priority for LIB waste management, at least without prior consideration of battery reuse. While conventional solid waste hierarchies place "reuse" above "recycling" in order of preference, LIB reuse has been far less studied. Yet clear benefits are promised: studies suggest that retired EV LIBs would still have 80% of their initial capacity intact (Nagpure et al. 2011; Marano et al. 2009; Zhang et al. 2011; Hoffart, 2008; Cready et al., 2003; Neubauer & Pesaran; 2011), and, directly recycling them without any consideration for reuse can forgo the benefit obtained from taking advantage of this remaining capacity.

Reuse of EV LIBs can theoretically have two forms – direct reuse in the application from which the battery was obtained (i.e., reuse in EVs) and cascaded use in a different and less demanding stationary application. LCA studies suggest that reuse of EV LIBs in stationary applications can provide environmental credits by avoiding the burden of manufacturing new battery packs for these end uses (Richa et al., 2015; Cicconi et al., 2012; Genikomsakis et al., 2013). For example, it was demonstrated in Chapter 3 that the extension of EV LIB life by use in a cascaded secondary application that displaces production of a lead acid battery system can result in Cumulative Energy Demand savings and Global Warming Potential reduction of as high as 69% and 68%, respectively (Richa et al., 2015). Additionally, reuse pathways would provide economic advantages for the EV user by potentially reducing the cost of EV batteries or EV lease payments, providing resale value for retired EV LIBs (Viswanathan et al., 2011; Neubauer and Pesaran, 2011; Neubauer et al., 2012; Williams and Lipman, 2010), and avoiding the cost of purchasing a new battery for the reuse application, as well as revenue generation potential for the utility sector (Neubauer and Pesaran, 2011; Narula et al., 2011; Heymans et al., 2014; Neubauer et al., 2012; Williams and Lipman, 2010; Viswanathan et al., 2011; Peterson et al., 2010, etc.).

While the general focus of recent literature has been on cascaded use of LIBs in stationary applications, reuse of these batteries in automobile application has not received much attention. A recent study by Saxena et al. (2015) suggests that LIBs can be used well below the 80% remaining energy capacity limit for less demanding daily travel needs of EV users or

extensive battery charging infrastructure, thus highlighting their automotive reuse possibilities. Even with the 80% LIB capacity fade limit, early vehicle failure (e.g. significant repairs, collisions) and EVs with battery replacement later in their useful life are likely to yield LIBs with high reuse potential as indicated in Chapter 2 (Richa et al., 2014) that could hypothetically be employed as replacement batteries for used EVs if technology and a market exists to support this system.

The last option for EV LIB waste management would be landfill disposal which is not expected to provide any environmental or economic benefit. The US EPA does not consider EV LIBs to be a major threat to environmental health as they do not usually contain toxic elements like lead, mercury or cadmium (Gaines and Cuenca, 2000). However, these batteries contain metals like lithium, aluminum, cobalt, manganese, nickel, and copper, which do have the potential to leach slowly into the soil, groundwater, and surface water if not disposed properly (Kang et al., 2013; Vimmerstedt, 1995). Similar to the case of electronic waste (Williams et al., 2008), risk of material leaching in well-managed sanitary landfills may be negligible, but the greater risk is loss of valuable materials, and the economic benefits of the reuse and recycling sectors should still avoid landfill of EV LIBs to promote a circular economy.

To delay or avoid landfill flows and tap into the potential benefits of reuse and recycling routes, the priority for managing the EV LIB waste stream is likely to follow a "waste management hierarchy". The EU Waste Framework Directive and the US EPA strongly advocate this circular thinking with a waste management hierarchy of prevention, reuse, recycling, other recovery (i.e. energy recovery) and disposal, with the highest priority waste management route resting at the top (European Parliament, 2008; US EPA, 2015b). In general, such a hierarchy depicts priorities from an environmental, and sometimes economic perspective for a variety of waste streams such as EOL electronics (Brandstotter et al., 2004), food waste (US EPA, 2016; Papargyropoulou et al., 2014; Glew et al., 2013), municipal solid waste (Cleary, 2009), packaging (Rossi et al., 2015), and construction waste (Batayneh et al.,2007), etc. but its validity is yet to be analyzed for LIBs.

Using concepts from circular economy and traditional waste management hierarchies, a theoretical waste management framework has been proposed here for EOL EV LIBs that includes reuse in EVs, cascaded use in stationary application, recycling and finally, landfill (Figure 4.1). A case study has been developed to examine the feasibility of this framework and

the hypothesis that the proposed approach, within known technical limits, will lead to improved environmental and economic benefits over recycling and/or disposal alone. This is a hypothetical, idealized case, because the goal is not to evaluate the impact of current practices for EV LIB waste management, but rather to analyze if the proposed waste management framework would be feasible from an eco-efficiency standpoint to achieve a circular economy. It should be noted that incineration was not included in the case study because of the dominance of landfill in the US with barely 13% of the total municipal solid waste combusted for energy recovery in year 2013 (US EPA, 2015a). It is recognized that the EOL management pathways for EV LIBs along the proposed hierarchy must also consider the existing and emerging policy landscape for these batteries and the products in which they are contained. Hence a policy analysis was also conducted to review the currently used mechanisms for battery waste management and identify the gaps in the current policy landscape to tackle future EV LIB outflows. Based on the gaps identified, the results of the case study were used to set out a roadmap for EV battery EOL management research and policy that considers the issues specific to these batteries as well as the goals of the circular economy (Ellen MacArthur Foundation, 2013; Geiser, 2001; Gregson et al., 2015; Ghisellini et al., 2016).

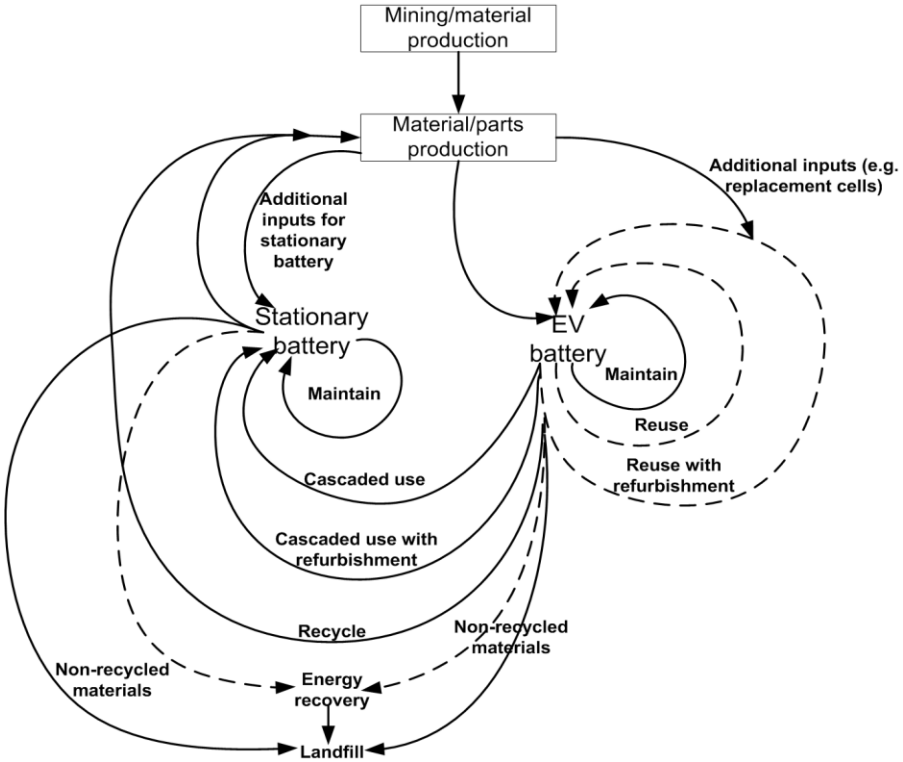


Figure 4.1 Theoretical waste management hierarchy for retired EV LIBs

4.2 Method

The different EOL management pathways of EV LIBs that were considered are reuse (in EV), cascaded use (in stationary applications), recycling and landfill (Figure 4.1). Both reuse and cascaded use pathways would include some level of testing and refurbishment to bring batteries back to a usable condition or prepare packs for new applications (Standridge and Corneal, 2014 and Richa et al., 2015).

For each of these pathways, the environmental metrics quantified by a life cycle approach were metal input, cumulative energy demand (CED), and eco-toxicity. These metrics were selected to demonstrate the environmental benefit or cost of the different waste management routes across the proposed hierarchy in terms of material resource, energy consumption and ecosystem quality, respectively. The net metal input in metric ton for different EOL pathways was estimated from the bill of materials of LIB cell and pack components, additional inputs and avoided battery systems in case of reuse/cascaded use of EV LIBs, and recycling efficiencies. The CED in megajoule (MJ) was calculated by using characterization factors and primary energy sources provided in SimaPro software CED calculation methodology. USEtox LCA impact assessment methodology was used to estimate the eco-toxicity impacts in terms of Comparative Toxic Unit for ecosystems (CTUe) (Rosenbaum et al., 2008). U.S. based electricity grid mix was used for electricity input data (ecoinvent Centre, 2010).The case study was applied to a theoretical stream of 1,000 LIB packs coming out of EV application in the future. This functional unit was selected to create results that could easily be scaled to an actual volume of waste packs, regardless of the time frame or total waste stream. Considering a conservative baseline of EV adoption, MFA results for year 2030 from Chapter 2 were normalized to 1,000 LIB packs to indicate a functional unit representing future annual EV LIB waste stream for a given year “n” (Refer to Appendix C Section S1). This waste stream was comprised of 25% BEV, 36% long range PHEV and 39% short range PHEV LIB packs (Richa et al., 2014)

Lithium Manganese Oxide (LMO) cathode chemistry was considered for EV LIBs due to its use as a constituent in blended cathode material of leading BEV and PHEV batteries (Lu et al., 2013) and has been used in the past to highlight the environmental impact of EV batteries (e.g. Notter et al., `2010; Dunn et al., 2012; Amarakoon et al., 2013). The energy capacity of these battery packs were based on the baseline scenario capacities from Chapter 2 (Richa et al.,

2014). The battery packs were modeled in Argonne National Laboratory BatPac tool (Nelson et al., 2011) resulting in an estimated waste stream of 160 mT of BEV and PHEV LIB packs for the chosen functional unit. The electric ranges of BEV, short range PHEV and long range PHEV were calculated in BatPac as 132 miles (39 kWh), 12 miles (18 kWh) and 50 miles (4.4 kWh), respectively. Additional comparisons were made on how some of the results of the case study would change if Lithium Nickel Manganese Cobalt Oxide (NMC) or Lithium Ferrous Phosphate (LFP) cathode or an equal proportion of all three (LMO, NMC and LFP) cathode types was used in EV LIBs, based on a similar approach by Richa et al. (2014) (For details, refer to Appendix C). Figure 4.2 illustrates the path of this EV LIB waste stream along waste management routes in the proposed hierarchy. These flows have been estimated from results in Chapter 2 and 3 (Richa et al., 2014; 2015) and are discussed in next sections. It is important to note that batteries entering into waste management in any given year "n" will cycle through multiple systems at various times in the future. These "cycles" (denoted by C1, C2 and C3) will each be separated by a time lag during which the battery is either in its reuse or cascaded use application or is recycled. For example, for year "n", 40% (by weight) of the EOL EV LIBs would not meet technical criteria required for reuse or cascaded use, and would therefore be sent for recycling in the same year (C1 recycling), while the rest of the waste would be recycled in later years after reuse and/or cascaded use, denoted by C2 and C3 recycling (Fig 4.2). Following sections discuss the approach employed for the eco-efficiency analysis of each of these EOL pathways for EV LIBs.

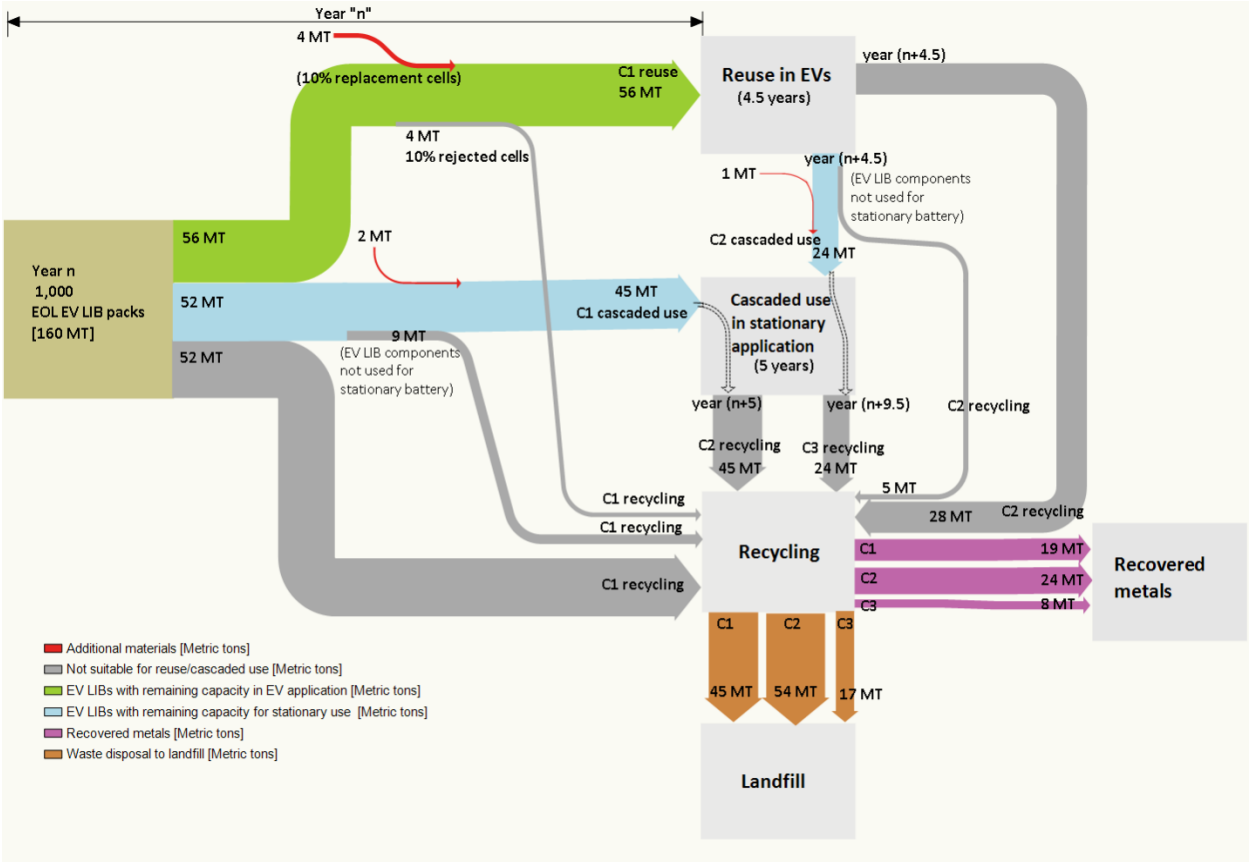


Figure 4.2 Diagrammatic flows of 1,000 EOL EV LIBs across different waste management routes. C1 denotes a given year "n" in which the battery waste flows out from first use in EVs and into the waste stream, C2 denotes year (n+4.5), to correspond with maximum expected lifespan for reuse in EVs, or (n+5), the maximum expected lifespan for cascaded use in stationary applications, and C3 denotes year (n+9.5), for batteries which were technically feasible to cycle through both direct and cascaded reuse.

4.2.1 Reuse in EV application

A maximum of 35% (by weight) of the EV LIB outflows in year n will have remaining capacity for use in EVs, having not yet reached the 80% capacity level due to early vehicle failure or crash or cases when an EV received a battery replacement and then reached the end of its life before the battery capacity is fully used ("lifespan mismatch" as described in Richa et al. (2014); Figure 4.2). An average of 9 years is considered as the "design lifespan" of LIBs for EV application (Standridge and Corneal, 2014 and Richa et al., 2014) corresponding to 3,285 cycles over the EV use phase of the LIB when cycled daily. Half of this design lifespan of the LIB is considered to be already spent in the first life EV use, leaving 4.5 years for reuse in EVs. This is a theoretical assumption, as direct reuse is not occurring in practice, to our knowledge, but a conceivable future scenario as EVs become more widely adopted. An underlying assumption for

this pathway is that 10% of the LIB cells would need to be replaced during refurbishment (Standridge and Corneal, 2014). Electrical performance is analyzed by using a conservative approach of testing whole battery packs by charging once to 70% battery capacity (Notter et al., 2010).

The net environmental impact of reusing EV LIB in vehicle application, $E_{reuse,\ EV}$ was calculated from the Avoided environmental impact of manufacturing new LIB pack (E_m), Design lifespan of EV LIB (l_d), Lifespan of EV LIB in reuse application (l_r), Impact of manufacturing replacement cells ($E_{m,cells}$), Impact of battery pack (or cells) testing (E_t), Impact of charge-discharge efficiency losses of refurbished LIB in EV application ($E_{c\text{-}d,\ reuse}$) and the Avoided impact of charge-discharge efficiency losses of new LIB in EV application ($E_{c\text{-}d,new}$):

$$E_{reuse,EV} = -(E_m/l_d) * (l_r) * E_{m,cells} + E_t + E_{c-d,reuse} - E_{c-d,new} \qquad (1)$$

Life cycle inventory (LCI) data for LIB pack production was obtained from Richa et al. (2015) and the efficiency loss calculations were based on Zackrisson et al. (2010). A direct correlation between capacity decay and battery charge-discharge efficiency was considered and after reuse in EVs, the capacity and efficiency was reduced to 80% (Ahmadi et al., 2014a; Richa et al., 2015) [Refer to Appendix C, Section S4].

The economic cost or benefit (in USD) of reusing old EV batteries for a second time in EV application ($V_{reuse,\ EV}$) was determined from the Avoided cost of buying new replacement LIB ($B_{LIB,new}$), Avoided resale value of the new EV battery at vehicle EOL ($S_{LIBused,\ new}$), Cost of buying a refurbished EV LIB ($B_{LIB,\ refurb}$) and Resale value of refurbished LIB at vehicle EOL($S_{LIBused,\ refurb}$) most likely for stationary applications:

$$V_{reuse,EV} = (B_{LIB,refurb} - S_{LIBused,refurb}) - (B_{LIB,new} - S_{LIBused,new}) \qquad (2)$$

Future new EV LIB cost ($125/kWh), refurbished battery buying price ($38/kWh) and used battery selling price ($20/kWh) was obtained for a low LIB cost scenario from Neubauer et al. (2012) based on similar future LIB cost estimates by U.S. Department of Energy (Howell et al., 2012; Neubauer and Pesaran, 2011).

4.2.2 Cascaded use in stationary applications

LIB packs from the EV LIB waste stream in year n would be employed for cascaded use in stationary application over two cycles, C1 (immediately on entering the waste stream) and C2 (after reuse in EVs). Some additional component input is required for refurbishing and assembling these systems (wiring, BMS, etc.) while some existing components may be discarded (Richa et al., 2015). Based on the expected cell failures during testing and technical limits of cascaded reuse, only 50% of the LIBs that outlived their usage capacity in EVs, were assumed to be feasible for stationary use, with a 5 year lifespan in stationary application (Richa et al., 2015).

The environmental benefit of cascaded use of retired EV LIBs was obtained from avoiding the production and use of lead-acid (PbA) batteries (450 kWh each) (Richa et al., 2015), the latter being the widely used technology for certain stationary and industrial purposes (Soloveichik, 2011) such as grid storage and off-grid renewable systems (Albright et al., 2012). The net environmental benefit from cascaded use of refurbished EV LIBs for stationary energy storage ($E_{reuse,\ stat}$) was calculated from the environmental impact of a refurbished EV LIB based stationary energy storage system ($E_{refurb,\ LIB}$) and the avoided impact of an equivalent functionality PbA battery system, E_{PbA}:

$$E_{reuse,stat} = E_{refurb,LIB} - E_{PbA} \quad (3)$$

LCA data for modeling environmental impact of production and use of PbA and cascaded use EV battery systems was obtained from Richa et al. (2015). The economic cost or benefit of the second use pathway ($V_{reuse,stat}$) was calculated from Refurbished battery buying price, $B_{LIB,refurb}$ (Neubauer et al., 2012) and valve regulated PbA (VRLA) battery buying price, $B_{PbA\ new}$ (Albright et al., 2012) for the utility sector:

$$V_{reuse,stat} = B_{LIB,refurb} - B_{PbA\ new} \quad (4)$$

4.2.3 Recycling

Recycling of EOL EV LIBs generated in year n and the additional material input for the reuse and cascaded use stages is likely to occur in three cycles C1, C2 and C3 separated by time lags (Figure 4.2). Net environmental impact of EV LIB recycling (E_{rec}) along these cycles was calculated considering equal proportion of the waste stream would be going to hydrometallurgical and pyrometallurgical recycling processes (Mudgal et al., 2011; Richa et al.,

2015) and was based on Environmental impact of recycling process ($E_{rec\ process}$) and the Avoided environmental impact due to material recovery ($E_{material\ recovery}$):

$$E_{rec} = E_{rec\ process} - E_{material\ recovery} \quad (5)$$

All metals in EV and stationary LIB packs were assumed to be recycled, however, owing to recycling inefficiencies, there will always be some fraction of these metals which would go to landfill. $E_{rec\ process}$ and $E_{material\ recovery}$ were calculated from LIB cell recycling and secondary metal production LCA data (Hischier et al., 2007; ecoinvent Centre, 2010; Fisher et al., 2006; Richa et al., 2015), and recycling efficiencies of metals contained in LIBs (Graedel, 2011; Sibley, 2011; Mantuano et al., 2006). Speculative recycling efficiency from bench scale recycling data for lithium and average recycling efficiency for manganese was used since these metals are not currently recovered from LIBs commercially (Gaines, 2014). Therefore, results represent the maximum theoretical value achievable from recycling.

The economic cost or benefit of EV LIB recycling (V_{rec}) was calculated from the Total cost of recycling operations (TC_{rec}) and Value of recovered materials ($V_{material\ recovery}$).

$$V_{rec} = TC_{rec} - V_{material\ recovery} \quad (6)$$

$V_{material\ recovery}$ was calculated using yearly average metals prices (USGS, 2015; Infomine, 2015) and recycling efficiencies. TC_{rec} was calculated based on methodology from Wang et al. (2014b) using fixed cost (1,000,000 $/year), maximum recycling capacity (34,000 mT annually), and variable cost (1,100 $/mT) for a recycling facility [Refer to Appendix C, Section S8].

4.2.4 Landfill

Some LIB materials, such as mixed plastics, graphite and electrolyte cannot be recycled due to lack of economic motivation or infrastructure (Richa et al., 2014). In addition, some metals will be lost from the value stream due to recycling inefficiencies. Thus, ultimate disposal options must still be considered for non-recoverable materials generated over the three temporal cycles. CED and indirect eco-toxicity impact of LIB materials landfilled was estimated using battery waste transportation and landfill operation LCI data (ecoinvent Centre, 2010). Results from laboratory based Toxicity Characteristic Leaching Procedure (TCLP) (see Appendix C,

Section S13) were used to estimate potential leaching of metals contained in LIBs disposed in landfills ("average leaching potential") [Table 4.1].

LIB metal	Average leachate concentration (mg/l)	Average Leaching potential (mg/kg)
Aluminum	131	11,000
Copper	1.61	200
Lithium	273	420,000
Manganese	335	110,000
Steel	13.7	3,100
Cobalt	15	36,000
Nickel	160	21,000

Table 4.1 Average landfill leaching potential of LIB metals

The TCLP represents an average leachate concentration over a moderate to long-term period of 3 to 10 years and for specific landfill design, waste composition and landfill water percolation characteristics (Frampton, 1998). These releases represent environmental loads, and their impacts on ecosystem quality were calculated by multiplying the total mass of a metal leached with the USEtox eco-toxicity characterization factor (in CTUe/kg) for that metal. Currently the eco-toxicity impacts of copper, manganese, iron, cobalt and nickel are characterized by USEtox.

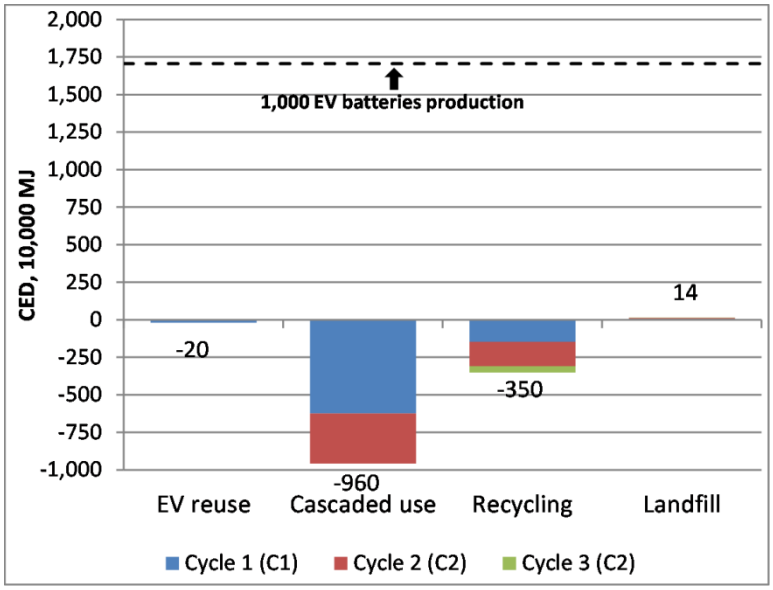
The implicit assumption is that non-recoverable materials from recycling operations will be sent to a municipal landfill, based on similar assumption for these batteries in E-waste in recent studies (Wang et al., 2014b; Kohler et al., 2008; Espinoza et al., 2014; Asari and Sakai, 2013). However it cannot be overruled that in many jurisdictions in future the EV LIB recycling residue maybe directed to industrial landfills and the metal leaching potential would vary in that case. An average landfill disposal cost of $1,170/mT of LIB waste was employed for estimating the economic cost of disposing waste LIBs in landfill. This included a collection fee of $1,120/mT (Wang et al., 2014b) and average landfill tipping fee for municipal solid waste in the US at $49.78/mT for year 2013 (US EPA, 2015a).

4.3 Results

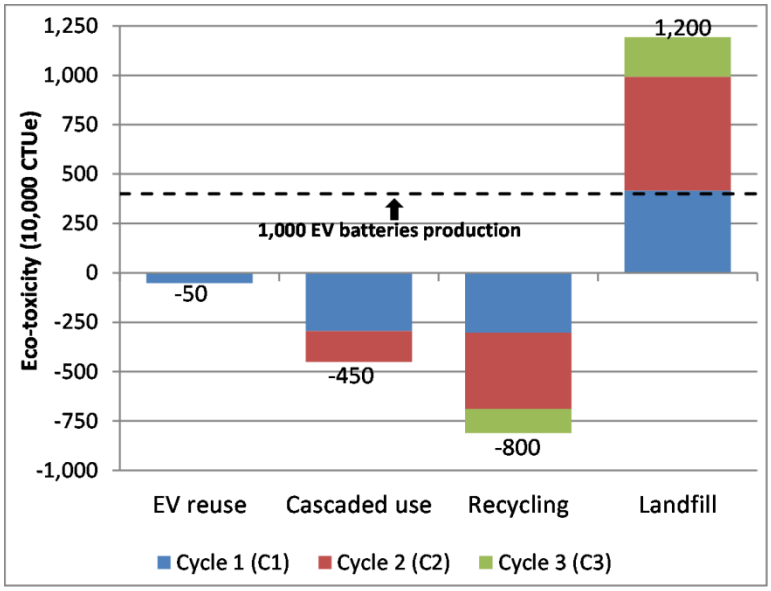
4.3.1 Eco-efficiency analysis of proposed LIB EOL hierarchy

The eco-efficiency analysis of the proposed EV LIB waste management hierarchy, including reuse, cascaded use, recycling and landfill, is discussed in the next sections. The

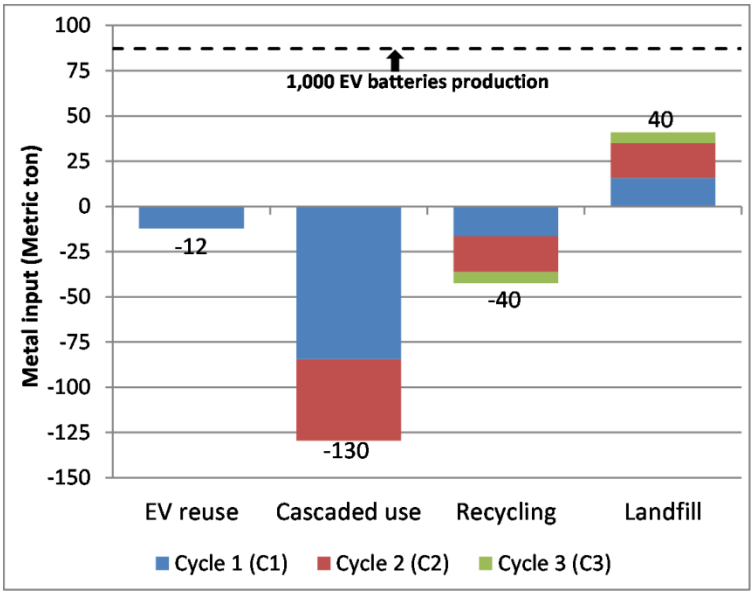
environmental results (Figure 4.3) are required to determine if a circular economy-inspired system will actually generate net environmental benefits or introduce unforeseen tradeoffs. The economic cost-benefit analysis for the case study is presented in Fig 4.4. Additionally, areas of uncertainties, which may cause deviations from these results, are discussed for specific cases.



(a)



(b)



(c)

Figure 4.3 Net environment impact of EV LIB EOL management routes for different fractions of the analyzed waste stream (1,000 LIB packs, during specific temporal cycles C1, C2, and C3) in terms of (a) CED, (b) eco-toxicity, and (c) metal input. The dotted lines represent impact due to production of these 1,000 LIB packs. A negative value of impacts denotes environmental benefit due to avoided production of new battery systems or avoided energy use in less-efficient displaced systems. These results showcase a circular economy inspired holistic approach of handling this waste stream and are not meant to be a comparison of different waste management routes. Figures showcasing the contributors to these net results in absolute terms are provided in Appendix C.

Reuse in EVs: Even though circular economy approaches would put product reuse at the top of the waste hierarchy, results actually show small if any benefit due to direct reuse of LIBs in EVs. LCA results (for e.g. in Chapter 3) have consistently pointed out the importance of the use phase in LIB life cycle associated with the declining round-trip efficiency and increasing energy losses as the battery ages (Richa et al., 2015; Ahmadi et al., 2014a,b). For reuse of BEV and high range PHEV battery pack in EVs, the avoided CED impact of LIB production far exceeded the CED of charge-discharge losses, replacement cells and LIB testing, resulting in a net benefit of 3,200 MJ and 73 MJ per pack, respectively. However, no CED benefit was observed for short range PHEV packs due to lower avoided CED value for these smaller packs, as compared to the CED impacts of electricity use and losses. Overall, a net CED benefit of 200,000 MJ was estimated from reusing the maximum feasible number of EV LIBs packs in automotive application (37.2% of the 1,000 packs entering the waste stream) (Figure 4.3 (a)). Benefits increase by 33% if short range PHEVs are excluded from the analysis. Since these benefits are primarily due to avoiding

new battery packs for EVs, LIB chemistries such as LFP with low CED impact for battery production would not provide a net CED benefit through this pathway (Refer to Appendix C).

If an aggressive testing procedure was employed for refurbishing EV LIBs for automotive reuse wherein individual LIB cells are tested for reuse capability, no CED benefit is expected in the reuse pathway across all EV LIB types. In fact, a net CED burden of 500,000 MJ would actually be created, due to energy efficiency losses plus energy required to test individual cells (See Appendix, Section S5). On the other hand, if it is assumed that energy efficiency of EV LIB does not decline with aging, the net CED benefit is expected to be as high as 2.6 million MJ. This represents a "hypothetical best case" since in general, efficiency degradation in remanufactured products may make them environmentally unfavorable in terms of energy consumption when compared to a new product for the same application (Gutowski et al., 2011).

Reuse of EV LIBs in vehicles can provide an eco-toxicity benefit per pack of 430 to 3,100 CTUe for short range PHEV to BEV battery, thus providing an overall eco-toxicity benefit of 500 thousand CTUe for the battery flows analyzed here (Fig 4.3 b). This benefit is obtained from avoiding the production of new LIB packs as replacement EV batteries which outweighs the potential eco-toxicity impact of electricity use even during aggressive battery testing. This pathway has the capability of avoiding approximately 12 mT of metals-primarily aluminum, manganese and copper (Fig 4.3 c).

The estimated economic benefit for the EV user from LIB reuse in vehicles ranges from USD 330 to USD 3,000 per pack for short range PHEV up to BEV battery. An overall economic gain of 500,000 USD is expected for the fraction of LIB packs reused in EVs from the analyzed waste stream (fig 4.4). These economic gains are tripled if high values of new EV LIB cost ($440/kWh), refurbished battery buying price ($132/kWh) and used battery selling price ($100/kWh) are considered (Neubauer et al., 2012).

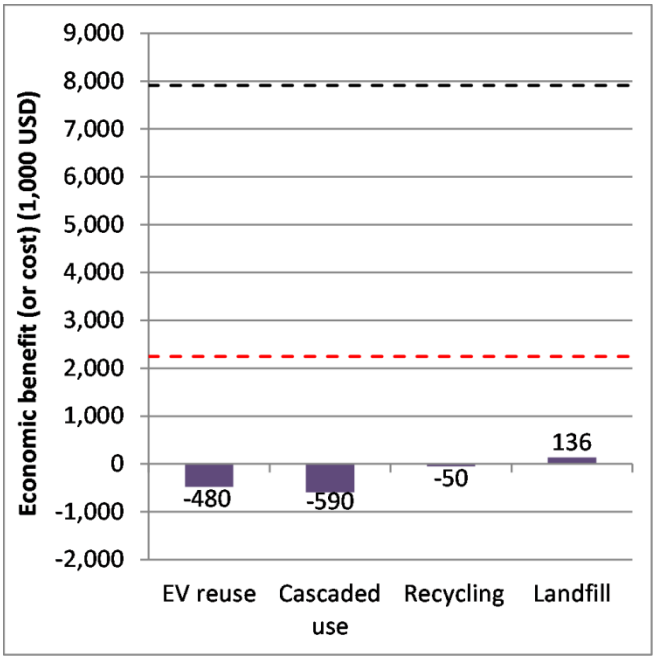


Figure 4.4 Net economic benefit or cost of EV LIB EOL waste management routes for different fractions of the analyzed waste stream (1,000 LIB packs). A negative value denotes economic savings while a positive value denotes economic cost. The dotted lines represent the "beginning of life" cost of the 1,000 EV LIBs in the waste stream for both high and low battery cost scenarios.

Cascaded use in stationary applications: For a stationary energy storage system operating for 5 years, a net CED and eco-toxicity benefit of 1,330 MJ/kWh and 626 CTUe/kWh can be obtained through cascaded use of retired EV LIBs and by avoiding the production and use of PbA battery systems. Translated into the overall benefit for the EV LIB waste flow functional unit analyzed here, a net CED and eco-toxicity benefit of 9.6 million MJ and 4.5 million CTUe can be obtained. These stationary battery systems would result in the avoidance of 130 mT of metal inputs, primarily by avoiding primary and secondary lead production. Adopting a waste management hierarchy wherein cascaded use is followed by recycling has the potential to recoup a significant fraction of energy used in the manufacturing of EVs along with their LIBs. For example, for a retired BEV battery, reuse in EV, cascaded use and finally, recycling can together recoup as high as 50% of the energy used to produce the vehicle and the battery and 40% of this gain is due to cascaded use (Refer to Appendix C, Section S12). Thus, the cascaded use strategy for retired EV LIBs can be instrumental in driving the circularity of the proposed system.

About 600,000 USD cost savings for the utility sector is estimated from installing these stationary energy storage systems with combined storage of 7,200 kWh (Figure 4.4). However, these estimates are based on low future LIB cost (Neubauer et al., 2012) and comparisons made

with Valve Regulated Lead Acid battery systems. There is immense uncertainty in future LIB prices and the type of PbA battery systems that would be replaced by these retired LIBs. When compared to flooded PbA battery systems, these cost savings would be reduced by 67% and in case of high future LIB cost scenario where the refurbished battery buying price could be as high as 132$/kWh, no economic benefit is expected from cascaded use.

Recycling: Recovery of metals from the original 1,000 EV LIB waste stream as well as any additional material input during reuse and cascaded use pathways provided CED and eco-toxicity credits of about 3.5 million MJ and 8 million CTUe, respectively. Using hydrometallurgical recycling provided 25% higher CED credits when compared to the pyrometallurgical process, primarily due to four times higher energy input for the latter (Fisher et al., 2006). For a mixed waste stream, comprising of equal fraction of LMO, NMC and LFP batteries, the CED savings can be slightly higher at 4 million MJ owing to slightly higher energy saving from recycling the latter two chemistries.

For the analyzed EV flows, pyrometallurgical recycling resulted in about 50 mT of avoided metals, while the metal recovery from the hydrometallurgical process was 29% less, leading to 40 mT of net avoided metals via both routes (Fig 4.3b). This is because the latter yields lithium, which constitutes 1-2% of the cells, while the pyrometallurgical process yields manganese comprising of 22-24% of the cells, with both metals exhibiting similar theoretical recovery efficiencies of about 50%. Direct recovery of LIB cathode through chemical re-lithiation to regain electrochemical performance at EOL has been demonstrated for LFP and LCO LIB cathodes and can provide the maximum energy savings, across all LIB material recovery pathways (Ganter et al., 2014; Dunn et al., 2012). However in this chapter, the more conservative hydrometallurgical and pyrometallurgical recycling processes were employed for the LMO chemistry as demonstrated by Fisher et al. (2006) for a European recycling facility, considering the uncertainty in future commercial recycling procedures.

A conservative cost (USD 1,100/mT variable cost) of recycling LIB pack materials results in overall recycling cost of 190,000 USD, for processing a total of 167 mT of materials. About 240 thousand USD in material value can be obtained across the three recycling cycles (~50 mT), resulting in net economic benefit of 50,000 USD (Figure 4.4). About 65% of this material value is derived from recycling LIB cells (~150 thousand USD). However, recycling

costs for LIBs are very uncertain- an average (USD 2,800/mT) vs. high end (USD 4,500/mT) cost of recycling operations (Wang et al., 2014b) change the net economic cost between 235,000 and 520,000 USD. For the conservative recycling cost scenario, a net benefit of 250,000 USD is expected if the waste stream was comprised entirely of NMC LIB packs, owing to high commodity value of cobalt (USGS, 2015). A mixed waste stream of LMO, NMC and LFP LIB packs would however result in negligible (~20,000 USD) material value (Refer to Appendix C).

Landfill: For all three cycles combined together, the quantity of waste LIB materials entering the landfill is expected to account for 70% of the total waste stream (of 1,000 EV LIB packs and additional material input during reuse and cascaded use). This landfill stream would constitute115 mT of non-recyclable materials such as mixed plastic, electrolyte, or graphite and recycling residues containing unrecovered metals. The cost of disposing this fraction of the waste LIB material was estimated at 136,000 USD (Fig 4.4). About 40 mT (35%) of this landfill stream was comprised of metals with 5% (~2 mT) of these metals potentially leaching in the landfill (Fig 4.5). Immense uncertainty exists over the amount of metals leaching from a landfill and these releases depend on numerous factors such as landfill age, control mechanisms, waste composition, water percolation, time dimension of leaching, metal degradation etc. (Olivetti et al., 2011; Rydh & Karlström, 2002). Slack et al. (2005) estimated 0.02% metal releases in landfill for non-battery waste, Fisher et al. assumed 5% leaching potential of heavy metals from spent batteries, while Rydh and Karlström (2002) assumed all metals from batteries to be released over an infinite time period, hence our results suggest a moderate leaching potential for LIBs.

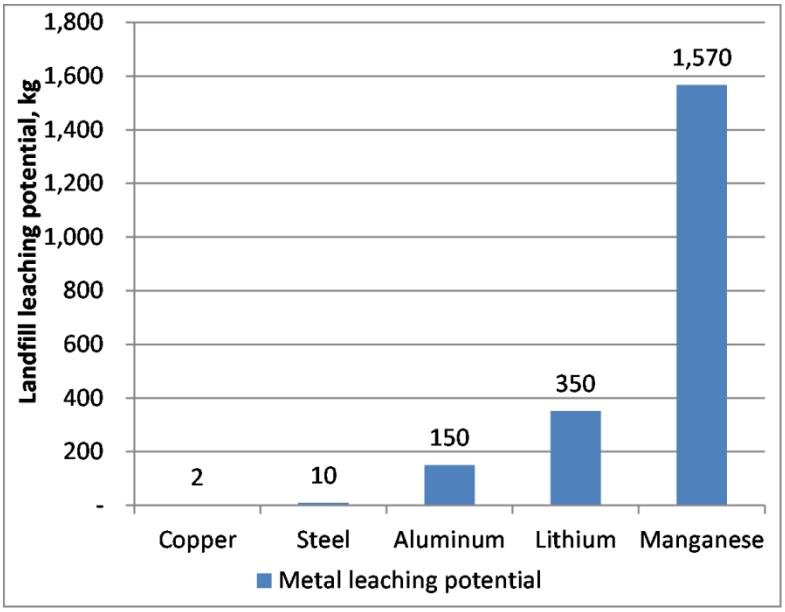


Figure 4.5 Landfill leaching potential of unrecovered metals for 115 mT of LIB waste in landfill

The US EPA has not set TCLP regulatory limits for these LIB metals, though the State of California has Total Threshold Leaching Concentration (TTLC) regulatory limits for cobalt, nickel and copper (Eurofins, 2012) which are heavy metals and are generally present in LIBs with mixed metal chemistries like NMC (Nelson et al., 2011).

The CED and "indirect" eco-toxicity impacts of landfill operation would be about 140,000 MJ and 14,000 CTUe, respectively. These impacts are primarily (>90%) due to waste transport to landfill, while energy and land use for landfill operation are negligible. "Direct" eco-toxicity implications of landfill leaching of metals from the analyzed landfill waste (Fig 4.5) would result in 12 million CTUe of eco-toxicity which far exceeds the upstream, indirect eco-toxicity impact for the bulk of the landfill waste. Use of nickel and cobalt based LIBs could aggravate the eco-toxicity impacts by an additional 13%, while a waste stream composed of only LFP batteries would reduce these impacts by 47% (Refer to Appendix C). Nevertheless, despite the widespread attention on eliminating battery landfill, these results underscore that a greater degree of toxicity impact can be avoided by battery reuse/cascaded use and recycle than would actually be generated if materials leach from landfills.

Consolidated Results: The consolidated results of this case study across various waste management routes were compared to the environmental impact of production and "beginning of

life" cost of the 1,000 EV LIBs in the analyzed waste stream (Fig 4.3 and 4.4). The CED and eco-toxicity values of LIB production were based on LCA study in Chapter 3, which considered a production mix of various metals used in battery manufacturing (Richa et al., 2015). Results suggest that the proposed waste management hierarchy would be able to recoup about 77% of the CED and 30% of the eco-toxicity impact of production of these batteries. In terms of metal inputs, the proposed system can avoid about 1.6 times of the metals used in LIB production, primarily owing to the cascaded use pathway which avoids large PbA battery systems. The proposed system can recover 12-44% of the initial cost of ownership of these batteries, however there is immense uncertainty in this domain.

4.3.2 Policy Analysis

The results of the case study indicate that a circular economy waste management hierarchy of reuse, cascaded use, recycling and ultimately landfill for EV LIBs would generally be consistent with expectations of benefits – with the exception of EV reuse, which is still technically unexplored and environmentally uncertain. However, given possibilities of economic barriers for the cascaded use and recycling routes, effective policy mechanisms are needed to encourage development of infrastructure to support this hierarchy, particularly when the economics may not favor implementation. Table 4.2 analyzes current policy mechanisms at the US and EU level that can impact EV LIB waste management in the future.

Life cycle stage	Initiative	Current policy that maybe extended for EV LIBs	Future mechanisms
Battery/Vehicle production	Material selection	**BD**: No restriction on LIB materials (i.e. considered non-hazardous) **ELV**: Limits use of hazardous substances in vehicles (doesn't explicitly address EV LIBs) **CA state** classifies LIBs hazardous due to excessive levels of cobalt, copper and nickel	•Can possibly regulate some LIB materials such as Co, Ni and nano materials •Chemistry standardization to avoid EOL sorting
	Design for EOL	**BD, ELV**: Appliances or vehicles to be designed to facilitate battery/component removal **BA**: Mandates ease of battery removal (Policy does not cover LIBs)	•Design principles for ease of disassembly, repair, refurbishment and recycling of EV LIBs as part of regulations
	Labeling or identification	**BD**: Labeling restricted to heavy material content (mercury, lead, cadmium) and landfill ban (crossed-out wheeled bin); capacity label non-mandatory **ELV**: material and component coding standards for identification **CA,MN**: Battery type (e.g. li-ion, NiCd, etc.) and note on recycling and safe disposal **BA**: Mandates labeling for battery type and recycling and safe disposal (but not LIBs)	•Specific labeling guidelines for LIBs: chemistry, capacity, LIB materials to facilitate sorting, remanufacturing and recycling. •Bar codes, RFID chips, specialized coloring for LIBs.
Use phase	Repair or maintenance	**EU Waste directive** defines "waste hierarchy" wherein waste prevention through product lifespan extension precedes other EOL management routes	No current end of life battery policy mandates repair/battery maintenance to extend life
Collection	Extended producer responsibility	**BD**: Collection financed by battery producers or third parties acting on their behalf **ELV**: EPR collection scheme applied to battery when collected with vehicle **CA,NY, MN**: retailer or battery manufacturer to provide for collection **Call2Recycle:** product stewardship program providing no-cost battery collection across U.S. and Canada funded by battery and product manufacturers	•Regulations to specify transfer of collection responsibility in case of reuse/cascaded use •In US states (NY, CA), collection programs are only for small, non-vehicular rechargeable batteries and may be expanded
Reuse/Cascaded use	Reuse/cascaded use provision	**BD**: Reuse or cascaded use not defined **ELV**: Reuse of vehicle component defined as use in same application; cascaded use in another application not defined; Mandates safe stripping operations, storage and testing to ensure suitability of vehicle component reuse (can include EV LIBs) **CA**: mentions "reuse"; not explicitly defined for LIBs	•Battery testing guidelines •Prioritize second applications based on techno-economic analysis from national labs in the U.S. •Economic incentives for reuse/cascaded use • Safety laws for large cascaded use installations
Recycling	Targets and process guidelines	**BD**: 50% recycling efficiency and rules for calculating the efficiency **ELV, BD**: Very brief guidelines for dismantling, storage and handling of batteries (e.g. electrolyte removal, removal of metals and plastic, sorting etc.) •No recycling or process efficiency targets in U.S.	•Rules related to worker safety and exposure •More specific dismantling manuals for EV LIBs •Addressing cross-contamination or developing mixed stream LIB recycling techniques •Economic incentives to promote recycling •Recycling efficiency improvement
Incineration and landfill	Prohibition	**BD**: Landfill and incineration prohibited **ELV**: Waste-to-energy and landfill of non-recycled vehicle components allowed **CA,NY,MN**: Landfill ban only (with ineffective or no penalty for non-compliance)	•Landfill ban to be extended to other US states •Awareness on landfill toxicity of LIB materials •Landfill tax, deposit-refund schemes, recycling incentives, differential taxation, etc.
Transport	Shipping guidelines (listed as Class 9 Miscellaneous hazardous material)	**BD**: Waste batteries exported for recycling to comply with waste shipment laws •LIB transport regulated by U.S. Department of Transportation, U.S. Hazardous Materials Regulations, International Civil Aviation Organization, International Air Transport Association, International Maritime Dangerous Goods which provide packaging, labeling shipping and fire hazard prevention instructions	•National and international regulations governing LIB transportation can be extended to waste LIBs and EOL battery management laws would mandate compliance •Specific guidelines for large size EOL EV LIBs

Table 4.2 Current policies governing LIB EOL management and future policy mechanisms. BD: EU Battery Directive; **ELV**: EU end-of-life vehicle directive; **NYS**: NY Rechargeable Battery Recycling Act (2010); **CA**: California's Rechargeable Battery Recycling Act (2006); **MN**: Minnesota Rechargeable Batteries and Products law (1994); **BA**: Federal Mercury-Containing and Rechargeable Battery Management Act of 1996

The EU Battery Directive (BD) which regulates the disposal of all battery types categorizes EV batteries as "industrial batteries" (European Commission, 2006) and is expected to include special provisions for EV LIB EOL management, unless an independent EV battery regulation is introduced. The EU EOL vehicle (ELV) directive provides guidelines for collection and EOL management of vehicle and their components (EU, 2000) and can be further expanded to include specific provisions for EOL EV LIBs. At the US level, the Mercury-Containing and Rechargeable Battery Management Act of 1996 (US GPO, 1996) (Battery Act [BA]) mandates that all recognized batteries (mercury-based, nickel-cadmium, and PbA) are to be considered hazardous waste and fall under the standards for Universal Waste Management. The Battery Act mandates guidelines for disposal of PHEV batteries but since the EPA Universal Waste Rule does not consider LIB materials to be hazardous, these batteries are excluded. However, the regulatory system could be adapted to include LIBs anticipating the sheer volume of retired EV LIBs. Only three US states have rechargeable battery waste management regulations that incorporate LIBs- California's Rechargeable Battery Recycling Act of 2006 (CA Code, 2006), New York State Rechargeable Battery Law (2010) and Minnesota Rechargeable Battery and Products Law of 1994 (MN PCA, 2015).

While the current policy landscape governing EOL batteries traverses through the entire battery life cycle (Table 4.2), mechanisms specific to EV LIBs would be required to handle the volume and complexity of this waste. Table 4.2 highlights future regulatory mechanisms that integrate the proposed waste management hierarchy with a life cycle approach, considering that product design and manufacturing are also integral stages in planning a circular system (Ellen MacArthur Foundation, 2013; Allwood et al., 2012; Allwood et al., 2011; Gregson et al., 2015; Bakker et al., 2014). Specific mechanisms are discussed across different stages of the LIB life cycle, including battery and vehicle production, battery use, EOL collection, reuse and cascaded use, recycling and landfill as well as transport of EOL batteries to enable their environmentally sound management:

Battery/Vehicle production: Initiatives for environmentally safe and convenient EOL management can be introduced at the point of LIB and vehicle production. Some examples include use of safe LIB materials and regulation of possible hazardous substances in EV LIBs, labeling requirements for easy identification and sorting of LIBs at their EOL as well as adopting

design-for-EOL (DfE) principles. DfE would enable easy removal of LIBs from EVs, facilitate battery disassembly, refurbishment, and material recovery (Arbabzadeh et al., 2016). These design principles can include modular battery pack design, use of compatible materials such as consistent grades of plastics, chemistry standardization, avoiding welded or soldered connections and minimizing the overall number of pack components (Chiodo, 2005). In this direction, The US Advanced Battery Consortium (USABC) is working on design-for-recycling guidelines for auto makers (USABC, 2014). The recommendations provided by USABC would require collaboration between battery designers and vehicle manufacturers as well as feedback from battery recyclers and vehicle dismantlers. The design elements proposed by the Consortium are specifically aimed to guide the battery-EV system designers and engineers. Some specific recommendations include (USABC, 2014):

1. Ease of removal: Suitable location of the battery in the vehicle with minimum number of similar fasteners for ease of removal from the vehicle, along with a "distributed system design" to allow easy separation from other system components such as BMS, etc.

2. Ease of disassembly of overall system: Assembly of the battery pack with minimum number of fasteners and of the same types, minimum use of adhesives, and avoiding incompatible adhesives and thermosets.

3. Safe removal: System design to allow for a complete and safe procedure to disconnect the high voltage contact points of the battery pack.

4. Chemistry identification: Easy identification of battery chemistry on the system as well as on individual modules, as per Society of Automotive Engineers (SAE) guidelines

5. Safe and easy removal of individual modules: Design of modules to enable maximum voltage to be less than 60V DC to reduce risks during disassembly. Moreover, enable modules to be easily disconnected without compromising on battery life

6. System Durability: Robust and long-lasting battery jacket (outer pack) design to enable reuse and refurbishing

7. Fluids management: Design of the system allowing easy removal of fluids such as electrolytes and coolants used for thermal management.

8. Ease of recycling: Battery pack using minimum number of different types of plastic materials and including appropriate labeling to separate plastics into different recycling streams. Additionally, allowing metals, plastics, electronics, and cells to be easily removed and sorted into designated recycling streams

Use phase: Currently none of the battery waste policies at the US or EU level emphasize maintenance or repair of batteries to extend their lifespan during the first use phase. Only the EU Waste Framework Directive has outlined a waste management hierarchy wherein waste prevention through product lifespan extension precedes reuse, recycling, energy recovery and disposal (European Parliament, 2008). Future waste policies can include specific guidelines towards ensuring a longer lifespan of LIBs in EV use owing to the expected lifespan mismatch between EVs and their batteries as indicated by Richa et al. (2014). Consumer awareness and education are likely to be incorporated into these guidelines.

Collection: At the EU level, effective extended producer responsibility (EPR) mechanisms are in place to ensure collection of waste batteries. The EU Battery Directive requires battery producers, or designated third parties, to finance collection, treatment and recycling of EOL batteries (European Commission, 2006). Under the ELV directive, EPR collection schemes apply only to vehicle components such as batteries and accumulators collected along with scrapped vehicles (EU, 2000). In the US, EPR mechanisms for LIB collection exist only in California, New York, and Minnesota wherein battery manufacturers or retailers must provide consumers with a free system for returning these batteries at EOL (CA Code, 2006; New York State Rechargeable Battery Law, 2010; MN PCA, 2015). Only Minnesota has set collection targets (90%, non-mandatory) for EOL rechargeable batteries and addresses EV LIBs by mandating EV and battery manufacturers to jointly manage EOL batteries

(MN PCA, 2015). New York and California laws and voluntary collection schemes in the US such as Call2Recycle are restricted to batteries from consumer electronics and exclude larger vehicle batteries (Call2Recycle, 2015). Hence US regulations need to be expanded in scope due to the large volume of retired EV LIBs expected. New regulations would be required to specify transfer of collection responsibility in case of reuse or cascaded use of EV LIBs.

Transport: LIBs are listed as Class 9 Miscellaneous hazardous material (Mikolajczak et al., 2011), and there are specific shipping, packaging and labeling guidelines for these batteries for transporting them domestically or internationally across various means of transport (Table 4.2). One major concern is the "thermal runaway" of LIB cells causing self-ignition leading to safety hazards (Webster, 2010). It is likely that different waste regulations that would govern EOL LIB management would mandate compliance to these guidelines and associated restrictions while transporting these batteries. For example, the EU Battery Directive mandates that waste batteries when exported for recycling should comply with waste shipment laws (European Commission, 2006). Additional safety requirements are expected for transport of retired EV LIBs owing to their large size, which may increase the cost of management of these batteries.

As per new regulations issued by the US Department of Transportation's [USDOT], Pipeline and Hazardous Materials Safety Administration [PHMSA], LIB packages transported by ground are required to be marked and labelled in the same way as for air shipments in accordance with International Civil Aviation Organization's standards (USDOT, 2014; Legal Information Institute, 2015). Currently, the transportation regulations provide specific guidelines for shipping LIBs or their cells and mandate limitations over the number of cells or batteries per package, package weight and packaging types for specific Watt-hour rating for these batteries (Legal Information Institute, 2015). Proper labeling in terms of Watt-hour ratings as well as lithium content and hazard communication are outlined by these regulations. There are additional requirements for shipping damaged or defective cells or batteries pertaining to labeling and packaging, which may apply for EOL EV LIBs as well, unless a separate regulation governs the latter. Currently, the USDOT prohibits the shipping of damaged, defective or spent LIBs meant for recycling by air transport. However, special permits are provided to LIB recyclers to transport used LIBs for recycling through highway, rail or vessel transport (USDOT, 2015). Such permits mandate United Nations (UN) specified packaging guidelines to prevent damage to cells

and short circuiting and require special labeling to indicate that the used batteries are meant for recycling. Moreover, some of the testing requirements and shipping documentation needed for transporting new LIBs are waived in special provisions of such permits. Additionally there are weight and Watt-hour limitations in such permits, for e.g. a 2015 USDOT permit for Retriev Technologies allows a maximum Watt-hour rating of 300 Wh per battery and limits the maximum gross weight of packages to 30 kg for transporting used LIBs for recycling (USDOT, 2015). Such permits are specific to recycling companies and as large sized EV LIBs are commercially recycled, new standards and guidelines may be needed to promote their safe transport.

Reuse and cascaded use: Considering the environmental benefits and possible cost savings for EV owners and the utility sector as demonstrated by the case study, it is prudent to consider these batteries for reuse and cascaded use prior to sending them to the next levels of the waste hierarchy. However, cascaded use is severely underrepresented in policies analyzed. While the ELV directive defines "reuse" of vehicle components for use in the same application, the more specific Battery Directive is only focused towards safe disposal and recycling of batteries (European Commission, 2006). 50% of currently in-use PHEV batteries in California can provide 850 MWh of energy storage capacity after retiring from vehicle application (Elkind, 2014). Waste management policies in the US do not explicitly address battery reuse or cascaded use, and second-life EV LIBs are currently not part of incentive programs or tax credits for grid or onsite energy storage systems in the US. To promote these reuse/cascaded use applications, future regulations can mandate battery testing protocols for specific applications and create economic incentives (such as tax rebates) for these pathways. Additionally, guidelines for automotive reuse and stationary application prioritization for second use LIBs based on the battery type (i.e. large BEV vs. small short range-PHEV battery; high energy vs. high power LIBs) and techno-economic assessments made by national laboratories in the US (Cready et al., 2003; Neubauer and Pesaran, 2011; Neubauer et al., 2012) can be provided. Such guidelines would enable diversion of these batteries towards appropriate extended life applications. Apart from regulatory deficiencies, possible regulatory barriers would govern siting of large stationary energy storage systems due to safety and environmental health concerns of second-life batteries (Elkind, 2014).

Recycling: Results suggest that recycling should be considered after exploring cascaded use options for EV LIBs. A well-established EPR network can incentivize recycling of EV LIBs that have undergone cascaded use, since some of the revenue from utility scale operations could bear the burden of their EOL management. The major focus of commercialized LIB recycling operations has been on cobalt recovery from consumer electronics batteries (Wang et al., 2014b). As suggested here, high costs of LIB recycling may provide small to no economic incentive from recycling non-cobalt chemistries such as LMO batteries. If the cells were composed of NMC cathode chemistry instead of LMO, the gross estimated value obtained from recycling these cells becomes more than two-fold (~340,000 USD) owing to high market value of cobalt and nickel (USGS, 2015). Hence, technical advancements to improve recycling processes, regulations to encourage EV LIB collection to promote economies of scale (Wang et al., 2014b) and incentives and rebates are needed to encourage recycling of low material value LIBs. The EU battery directive has recycling efficiency mandates of 50% for batteries including LIBs but no legislation within US provides process targets. Owing to expected variability in chemistry and composition of EV LIBs, their recycling will need to avoid cross contamination or develop recycling procedures to process different LIB chemistries simultaneously. The Society of Automotive Engineers in the U.S. and EUROBAT in the E.U. have established active working groups to develop solutions for battery labeling and to prevent cross contamination in LIB recycling streams (Gaines, 2014).

Apart from policy deficiencies, LIB recycling can face regulatory hurdles due to workplace exposure to LIB materials during battery disassembly and shredding (Wang et al., 2015). The ELV directive provides generic guidelines for dismantling, storing and handling vehicle batteries but does not address issues specific to LIBs. Majority of LIB cathode and electrolyte materials have OSHA hazards associated with them (Vimmerstedt et. al., 1995) which may require workplace regulations for LIB recycling facilities, further raising the cost of recycling operations.

Landfill: While the share of EV LIB waste ultimately reaching the landfill was estimated at 70% of the entire waste stream, the cost of landfill was estimated to be less than even the most conservative cost of recovering 30% of these materials. Preliminary TCLP analysis of bulk of LIB metals (Table 4.1) suggests that metal concentration in LIB leachate could exceed the US

Primary and Secondary Drinking Water Standards (US EPA, 2009), as well as the EU Drinking Water Directive and World Health Organization's guideline limits (European Union, 1998; WHO, 2008). If the landfill leachate from LIBs were to contaminate the groundwater, it could pose a potential threat to human health and environment.

Hence, appropriate landfill tax and widespread and stringent landfill bans are required. Based on the regional tipping fee and the expected EV LIB waste disposal for specific states, regional variability is expected in LIB landfill disposal expenses. Differential landfill tax can be introduced based on increasing LIB waste tonnage for a given region. Economic instruments to encourage LIB recycling, such as deposit-refund schemes (Walls, 2006), or incentivizing recycling can prevent non-compliance to landfill bans. Additionally, improved recycling efficiencies of LIB metals and recycling currently non-recovered LIB components such as electrolyte, graphite, and plastics (Richa et al., 2014) is needed.

4.4 Conclusion

A circular economy-centric waste management hierarchy can be effective in managing the EV LIB waste stream in future, but uncertainties exist over its eco-efficiency. Both EV LIB reuse and cascaded use have potential for providing environmental benefits. However such benefits rely significantly on LIB size, testing procedures, and the incumbent battery systems that used LIBs would displace. While there is potential for these extended life pathways to provide EV owners and the utility sector with cost-effective batteries, such cost savings would depend on future prices of new and old EV LIBs, stationary energy systems that these batteries are replacing, and regulatory barriers due to environmental health and safety concerns. Results indicate that a "closed" circularity of LIBs in the same EV product is less desirable than a more "open" loop cascaded use into stationary applications, which is contrary to typical results expected from a circular economy model. The expected benefit from cascaded use pathways of EV LIBs calls for policies and economic incentives to preferentially promote cascaded use over recycling across a waste management hierarchy. Additionally, environmental benefit from LIB recycling should stimulate policies to promote profitable recycling operations and avoid battery landfill through extensive collection programs, improved recovery efficiencies and economic incentives. Additional policy may be needed to promote worker health and safety and ease of recycling.

Considering the large fraction of EV LIB material flows into landfill, policy mechanisms are needed to implement effective and widespread landfill bans and stringent landfill penalties to improve the circularity of the proposed system. The proposed waste management hierarchy can be instrumental in diminishing eco-toxicity impacts over this extended EOL phase of EV LIBs, considering reuse and cascaded reuse of EV LIBs, followed by recycling can negate the eco-toxicity burden of unavoidable metal flows into landfill. A comprehensive life cycle based approach that additionally considers design for EOL, battery maintenance, collection and safe transport of EOL EV LIBs would prove effective in developing policy instruments that promote a circular waste management system.

CHAPTER 5: CONCLUSION AND MAJOR CONTRIBUTIONS

The adoption of electric vehicle has been gradually increasing with time (Inside EVs, 2015), and this trend shows promise towards reducing dependence on fossil based transportation fuels. However, increasing adoption of EVs in future would be accompanied with large scale production of LIBs to power these vehicles and a waste management problem when these batteries reach their end-of-life in vehicle application. Hence, sustainable waste management strategies and related policy framework is required to handle this battery waste stream in future.

To gain a better understanding of the scale at which EOL battery recycling infrastructure must be developed in future, Chapter 2 demonstrated a future oriented top-down material flow analysis (MFA) to estimate the volume of EOL EV LIBs generated in the near and long term future. Owing to the potential "lifespan mismatch" between battery packs and the vehicles in which they are used, both reuse and recycling potential exists for these batteries in future. In fact, there is a possibility that 37% to 43% of LIBs will be reused in vehicle applications itself. The commodity value of materials contained in the future EV LIB waste stream will vary with cathode chemistry composition of the stream. Cost efficient recycling processes will be needed for currently non-recycled materials like lithium and manganese as automotive manufacturers are transitioning to low cost EV LIBs. In terms of recycling, the actual economic value of EV battery recycling would depend on the LIB collection rates and recovery rates of the various materials present in the stream. Moreover, safe disposal of low value battery materials will be required owing to their large volumes in the waste stream. Due to the high tonnage and material variability expected in the LIB waste stream in future, LIB recycling infrastructure must be able to handle the scale as well as complexity of this waste stream.

Chapter 2 additionally demonstrated the uncertainties encountered in conducting dynamic MFA of emerging technologies which is inherent in the emerging field of industrial ecology. For the MFA model it is assumed that the material composition of the four battery technologies remain the same over the temporal boundary of the analysis. Future work can look at how the flows can possibly vary, as the specific energy of LIBs improves with time, thus possibly reducing the material input per battery pack over the next decades. Additionally, inclusion of novel battery technologies in the model, such a lithium-air and LIBs containing nano materials can provide a more enhanced picture of the impact of technological progress on the battery

outflows from EOL EV LIBs. In this Chapter, a hypothetical lifespan distribution of EV LIBs was assumed owing to lack of current data, while the lifespan of EVs was considered to be constant. Currently both the batteries and EVs are in their early stages of adoption. As more data becomes available in terms of aging and obsolescence of the two different components of this dual product system, more refined estimates of future battery outflows can be obtained. While it is acknowledged that the forecasts in Chapter 2 exhibit significant uncertainty at early stages of technological deployment, it provides the very first estimate of waste battery outflows as well as a robust modeling framework for extended analyses as these data uncertainties are resolved in future. The MFA framework adopted in Chapter 2 additionally provides a model case study for analyzing waste outflows for other dual-product systems similar to the EV-LIB system.

Chapter 3 demonstrated that cascaded use of retired EV LIBs in stationary application exhibits environmental trade-offs due to impact of LIB refurbishment and charge-discharge efficiency losses and the credits obtained from avoiding the manufacturing and use of incumbent PbA batteries. The environmental feasibility of such second use systems would depend on reuse feasibility of LIB cells and their reliability to provide competitive service life in stationary applications. This is owing to the direct effect of these technical parameters on the refurbishment stage. While low cell conversion for stationary application implies more LIBs would be transported and tested for capacity and electrical performance, a small service life of refurbished cells would require multiple battery replacements for the stationary energy storage system. In general, results in Chapter 3 demonstrate that EV LIB reuse in stationary application has the potential for dual benefit – both from the perspective of offsetting initial manufacturing impacts by extending the lifespan as well as avoiding production and use of a less-efficient PbA battery. However, these benefits can diminish when very few of the initial battery cells and modules could be reused and where reliability is low (e.g., 1 year or less lifespan) in the secondary application.

It is concluded that reuse decisions and diversion of EV LIBs towards suitable stationary applications can be based on life cycle-centric studies. For example, retired EV LIBs with high cell conversion rate but short stationary service life would still prove environmentally feasible to employ in less intensive stationary storage or ad-hoc applications. These can include less frequently needed (~twice per year) power back up systems such as those for telecommunication applications (Cready et al., 2003). In such sporadic use applications, the remaining LIB cycles

can be spread over a larger time frame, which could raise their preference over current PbA batteries. This preference would be additionally substantiated owing to a much higher temperature sensitivity of PbA batteries in these applications which accelerates their aging process and reduces their calendar life (Cready et al., 2003). Chapter 3 further suggests that technical feasibility must be evaluated, particularly with respect to the ability to rapidly analyze the reliability and remaining cycle life of EV LIB cells, modules, or packs for refurbishment and reuse in secondary applications. While LIB lifespan extension through stationary reuse would delay the entry of EOL EV LIBs into the waste stream, a sustainable market for used EV batteries would rely on economic motivation, technical feasibility and stakeholders' acceptability- factors which need to be further analyzed.

Apart from outlining these imminent issues, Chapter 3 strengthens the knowledge of battery LCA since inclusion of a second use phase in LIB lifespan provides a comprehensive assessment of their environmental implications in future in contrast to previous studies (Majeau-Bettez *et al*., 2011; Notter *et al.*, 2010; Zackrisson *et al*., 2010; Samaras and Meisterling, 2008; Ishihara et al., 2002; Hawkins et al., 2013; Matheys et al., 2009, Ellingsen et al., 2013) which were limited to the production and EV use of these batteries. A major modeling challenge addressed was the allocation of impacts between the EV and stationary use phases of EV LIBs. These allocation strategies can be applied to other LCA models investigating product use cascades, which have till now received limited attention in the LCA realm. The choice of allocation method will guide LCA outcome and can pose a major challenge towards environmental assessment of these systems. For the EV LIB cascaded use system, the selection of allocation method can likely depend on the battery ownership model and responsibility for their EOL management across various stakeholders involved such as battery material producers, battery producers, vehicle producers, vehicle dealers, vehicle treatment operators, and utility providers.

While a cascaded used EV LIB system was compared to lead acid battery in Chapter 3, the environmental feasibility of these systems additionally needs to be analyzed in comparison to other prominent and upcoming battery technologies for utility applications, particularly redox flow batteries and high temperature sodium batteries. Additional research is required to understand how retired EV LIBs compare to new LIBs for stationary applications, in terms of cost, performance as well as the net environmental impacts. While a generic stationary

application was analyzed for environmental feasibility of EV LIBs, future work can possibly compare the environmental impacts of the cascaded use route across a wide range of grid-based and off-grid stationary applications, including high power and high energy applications such as power back-up, renewable firming, load leveling, transmission support etc. as elaborated in Cready et al. (2003). Each of these applications has different depth-of-discharge of operation, frequency of use and system lifespan and hence their comparison is a challenge, though necessary from a feasibility perspective.

In Chapter 4, an eco-efficiency analysis was conducted to analyze the possible environmental and economic costs and benefits of different EOL management routes for EV LIBs, along a circular economy inspired waste management hierarchy. Results indicated that if technology and markets support reuse of LIBs in used EVs, the net benefit would be 200,000 MJ of recouped cumulative energy demand (CED), which is equivalent to avoiding the production of 11 new plug-in EV batteries (18 kWh capacity each). Avoiding production of replacement LIBs for EV applications also promises benefit of reduced eco-toxicity and metal depletion. These benefits are magnified almost ten-fold when retired EV LIBs are cascaded in a second use application for stationary energy storage, thereby replacing the need to produce and use less efficient lead-acid batteries. Reuse and cascaded use routes can likely provide EV owners and the utility sector with cost savings of as high as 1.7 million and 600,000 USD, respectively for the analyzed waste stream. Since, both reuse of EV LIBs in vehicle application and cascaded use in stationary energy storage provide scope for environmental impact reduction and cost savings, recycling should not be the first option for EOL management route for these batteries until their reuse and second use potential have been explored. However, the environmental benefit from the reuse and second use pathway are likely to depend on: (a) the refurbishment procedure-whether the feasibility analysis of these LIBs employ testing of individual cells or testing of whole packs, and (b) the avoided battery system that these reuse and cascaded use systems are being compared to while calculating the environmental benefits. Similar uncertainty exists in terms of the economic gains for these EOL routes owing to speculation of future new and used EV LIB prices and the cost of the avoided battery system being compared (e.g. lead acid battery in the case study in Chapter 4).

For the waste stream composed of Lithium Manganese Oxide EV LIBs analyzed in Chapter 4, recycling pathway provided little (240,000 USD for the analyzed waste flow) to no

economic benefit owing to the high costs of recycling operations. However, recycling of these batteries for metal recovery has tremendous potential for environmental savings by avoiding the mining and production of primary metals. There is no environmental or economic benefit from the landfill of EV LIBs, hence landfill disposal should be the last resort during their EOL management. In fact, the landfill pathway is expected to account for 69% by mass of the EV LIB waste flows in a given year- both due to recycling inefficiencies of metals and also due to the large fraction of low value, non-recyclable materials in the stream as also exhibited in Chapter 2.

Owing to the expected benefits-environmental or economic or both for reuse, cascading use and recycling of EOL EV LIBs, a waste management hierarchy needs to be adopted for sustainable management of this waste stream in future. Such a hierarchy would enable all possible beneficial waste management strategies to be explored before sending these materials for waste disposal, thus attempting to close the loop for a large fraction of the waste flows. However, the current policy landscape does not encourage reuse or cascaded use of these batteries and have ineffective regulations for encouraging material recovery and compliance to landfill bans. The expected benefit from reuse and cascaded use pathways of EV LIBs can stimulate future policies aimed towards promoting battery design-for-EOL, collection for reuse and cascaded use, reliable and cost-efficient testing procedures as well as economically and technically favorable second use pathways. In case of environmentally favorable but economically infeasible second use routes, economic incentives maybe introduced to preferentially promote the second use over recycling across a waste management hierarchy.

It is expected that the large environmental benefit from LIB recycling should stimulate policies to promote widespread EV LIB collection programs, profitable recycling operations and technical advancements to achieve higher recycling rates and efficiencies. These policy mechanisms would be specifically required for non-cobalt containing, low material value EV LIBs, such as those composed of Lithium Manganese Oxide and Lithium Iron Phosphate. Currently, in US only the states of California, New York and Minnesota have imposed landfill bans for LIBs. However, most battery collection programs in US exclude large sized vehicle batteries, which can lead to violation of the lenient landfill disposal bans. The high volume of EV LIB landfill waste accompanied with eco-toxicity impacts of metals leaching in landfill would call for policy mechanisms to implement effective and widespread landfill bans and stringent penalties based on volume of landfill waste generated. The results of the eco-efficiency analysis

and the wide gaps in the current policy landscape demand a more robust EV LIB waste management policy framework in future that can likely be based on a life cycle based model. Moreover, these findings underscore the importance of life cycle and eco-efficiency analysis to develop proactive policies for improving the "circularity" of the proposed system.

The circular economy inspired waste management hierarchy model presented in chapter 4 can be further expanded in future work to develop a decisions analysis tool, where the different inputs to the model can be changed to understand the eco-efficiency of the proposed system under a range of scenarios. Currently immense uncertainty exists about the cost of new and refurbished EV LIBs in the future as well as pertaining to the cost of operation of future recycling facilities. Similarly, the battery collection and landfill operation costs can vary within different states in the US. Additionally, for an analyzed waste stream of EV LIBs, the chemistry mix is highly uncertain. The dynamic model can be used to capture these uncertainties and variability. Apart from CED, metal depletion and eco-toxicity a set of other environmental metrics can be incorporated in the model, depending on what the stakeholders deem important for their analysis and decision making.

APPENDIX A

S1. List of parameters

Parameter	Description
t	A given year. For EV battery inflow, t= 2009 to 2035; For EV battery outflows, t=2015 to 2040
l	EV LIB lifespans (Scenario based): Refer to figure 2.3 in Chapter 2
P_l	Percentage of EV LIBs sold in a given year to have a lifespan of l years (Scenario based): Refer to Figure 2.3 in Chapter 2
K	EV LIB inflow (i.e. LIB input into EVs)
i	EV type: BEV, PHEV10, PHEV40, and HEV
S	Sales of new EVs that use LIBs
W	Non-EOL EVs requiring a replacement LIB
B	Number of LIB packs in EV battery waste stream
$PE_{i,t}$	Percentage of waste LIB packs belonging to EV type i in year t: ($B_{i,t}/B_t$)
j	LIB cathode chemistry: Lithium cobalt oxide ($LiCoO_2$), lithium manganese oxide ($LiMn_2O_4$), lithium iron phosphate ($LiFePO_4$) and lithium nickel cobalt manganese (NCM) oxide
PC_j	Percentage of LIBs of cathode chemistry j in EV battery waste stream: 10% $LiCoO_2$, 30% $LiMn_2O_4$, 30% $LiFePO_4$, and 30% NCM
D	Number of cells per EV LIB pack
Epack	EV LIB pack energy storage (Watt-hour)
R	Electric range of EV (miles): 100 miles for BEV; 40 miles for PHEV40; 10 miles for PHEV10 and 4 miles for HEV
C	EV energy consumption rate (Watt-hour/mile)
η	Percent efficiency of EV LIB
A	Percent available energy of the total EV LIB energy
Ecell	Lithium ion cell energy storage (Watt-hour)
N	Number of lithium ion cells in EV battery waste stream
y	Materials in lithium ion cell: Aluminum, cobalt, copper, lithium, manganese, nickel, steel, iron, graphite, carbon black, binder, plastic, electrolyte, others
m	Material mass in lithium ion cell
MI	Material mass of EV LIB inflows
MO	Material mass of EV LIB outflows
g	EOL EV battery type based on reuse potential: Type 1 and Type 2 EOL LIBs

Table S1.1 List of MFA parameters

S2. Lithium ion battery use in hybrid electric vehicles

Estimates from Jobin et al. (2009) for a Credit Suisse report were applied to the scenarios, as their study provided both conservative as well as optimistic estimates for hybrid electric vehicle (HEV) lithium ion battery (LIB) adoption in comparison to HEV Nickel metal hydride (NiMH) battery usage.

S2.1 Credit Suisse bottom-up HEV LIB penetration estimates

Year	LIB HEV	(LiB+NiMH HEV)	% HEVs using LIBs= LIB HEV/(LIB+NiMH HEV)
2009	0%	100%	0%
2010	4%	98%	4%
2011	11%	95%	12%
2012	18%	93%	19%
2013	31%	91%	34%
2014	39%	89%	44%
2015	45%	86%	52%
2016	50%	83%	60%
2017	52%	80%	65%
2018	53%	76%	70%
2019	53%	72%	74%
2020	51%	68%	75%

Table S2.1 Market Volume by battery technology (LIB+NiMH) (Adapted from Jobin et al., 2009)

The high and baseline scenarios employed Credit Suisse bottom-up estimates which are more optimistic towards HEV lithium ion battery adoption. These estimates were available till year 2020. Even though, these estimates suggest market share of different electric vehicle (EV) technologies in the future, they have only been used in our study as a basis for calculating the percentage of HEVs using lithium ion battery technology as opposed to nickel metal hydride technology. For market share of different EV technologies, the Energy Information Administration (EIA) Annual Energy Outlook estimates were employed. It was additionally assumed that all HEVs would use lithium ion batteries by year 2015 and 2025 in the high and baseline scenarios, respectively. The HEV lithium ion battery adoption rates for year 2014 for high scenario and for years 2021-2024 for the baseline scenario were estimated by means of linear interpolation.

S2.2 Credit Suisse top-down HEV LIB penetration estimates

% HEVs using Li-ion (vs. NiMH)	
2009	1%
2015	27%
2020	62%
2025	72%
2030	82%

Table S2.2 Percent HEVs using LIBs

The Credit Suisse top-down estimate for HEV lithium ion battery adoption were used for the low scenario, with the additional assumption of hundred percent lithium ion battery adoption by HEVs in the year 2032. This assumption was based on the forecasted HEV adoption rate using the forecast function in excel as the Credit Suisse top-down estimates were only available till

2030. Moreover, as this data was available for every five year, the data gaps were estimated by means of linear interpolation.

The percent of HEVs using lithium ion batteries for a given year was multiplied with the HEV sales forecast for that year to yield the number of HEVs using lithium ion batteries in that year.

S3. Cell energy storage

Energy storage of 18650 (cylindrical) form factor lithium ion cells for the four battery chemistries was obtained as the product of cell capacity and the nominal or average cell voltage. The cell capacity (mAh) was estimated as the product of the cathode mass and the specific capacity (mAh/g) of lithium ion cells for each of the four cell chemistries considered in the model. The specific capacity of the lithium ion cells was obtained from Dahn and Erlich (2011). The cathode mass of each of the cell types was estimated from their respective bill of materials. A sample calculation for estimation of cathode mass of cylindrical $LiFePO_4$ cells is shown.

For $LiFePO_4$:
Li - 1 mole = 6.94g
Fe - 1 mole = 55.845g
P - 1mole = 30.974g
O - 1mole - 15.99g * 4 moles = 63.96g

Total molar mass of $LiFePO_4$ = 157.719g

So the mass percentage of each element in the structure would be:

Li - 6.94g/157.719g = 4.4%
Fe - 55.845g/157.719g = 35.4%
P - 30.974g/157.719g = 19.6%
O - 63.96g/157.719g = 40.6%

From the bill of materials of $LiFePO_4$ cells,
Mass of lithium in a 18650 cell = 0.51 g
Mass of iron in a 18650 cell = 4.11
Hence mass of cathode active material of $LiFePO_4$ cell = (0.51/0.044) g or (4.11/0.354) g
= ~ 11.60 grams
In a similar way, the mass of the cathode active materials of cylindrical lithium ion cells of other chemistries was calculated. The following table describes the calculation of cell energy from cathode mass, specific capacity and cell voltage of lithium ion cells:

Cathode Chemistry	Cathode mass (g)	Specific Capacity (mAh/g)	Cell Capacity (Ah)	Average/Nominal Cell Voltage (V)	Cell Energy, Ec_i (Wh)
Lithium Cobalt Oxide ($LiCoO_2$)	13.25	155	2.05	3.84 (Howard & Spotnitz, 2007)	7.89
Lithium Manganese Oxide ($LiMn_2O_4$)	15	115 (Average)	1.72	3.86 (Howard & Spotnitz, 2007)	6.65
Lithium Iron Phosphate ($LiFePO_4$)	11.60	160	1.86	3.25 (Burke & Miller, 2009)	6.04
Lithium Nickel Cobalt Manganese Oxide ($Li(NiMnCo)O_2$)	14.65	160 (Average)	2.34	3.60 (Burke & Miller, 2009)	8.44

Table S3.1 Cell energy of 18650 form factor lithium ion cells

As a result of the assumptions and calculations documented here in Section S3 and in the Chapter 2, the total number of cells for each scenario and each type of battery pack was determined.

Scenario	**Cathode Chemistry**	**No. of cells per BEV battery pack**	**No. of cells per PHEV10 battery pack**	**No. of cells per PHEV40 battery pack**	**No. of cells per HEV battery pack**
Low	$LiCoO_2$	3,700	420	1,700	450
	$LiMn_2O_4$	4,400	500	2000	530
	$LiFePO_4$	4,900	550	2,200	580
	$Li(NiMnCo)O_2$	3,500	390	1,600	420
Baseline	$LiCoO_2$	5000	560	2,300	680
	$LiMn_2O_4$	5,900	670	2,700	800
	$LiFePO_4$	6,500	740	2,900	890
	$Li(NiMnCo)O_2$	4,600	530	2,100	630
High	$LiCoO_2$	6,500	750	3,000	1,000
	$LiMn_2O_4$	7,700	890	3,500	1,200
	$LiFePO_4$	8,500	980	3,900	1,400
	$Li(NiMnCo)O_2$	6,100	700	2,800	980

Table S3.2 Number of cells per EV lithium-ion battery pack

S4. Bill of materials of lithium ion cells (18650 form factor)

The bill of materials of 18650 form factor lithium ion cells was obtained from Wang et al. (2014a) for the 4 cathode chemistries.

	$LiCoO_2$ (LCO)	$LiMn_2O_4$ (LMO)	$LiFePO_4$ (LFP)	$Li(NiMnCo)O_2$ (NCM)
Material, *y* (grams)				
Aluminum	2.40	0.50	2.76	2.22
Cobalt	7.97	0.00	0.00	4.08
Copper	3.36	0.50	3.45	3.29
Lithium	0.94	0.69	0.51	0.48
Manganese	0.00	9.11	0.00	3.81
Nickel	0.56	0.00	0.00	4.07
Steel	7.60	7.35	14.18	7.31
Iron	0.00	0.00	4.11	0.00
Graphite	10.64	15.00	5.50	7.26
Carbon black	2.78	0.00	0.99	2.55
LiPF6	1.71	0.00	0.49	2.05
Ethylene Carbonate (EC)	0.43	0.15	3.42	0.51
Binders	1.11	0.00	0.39	1.02
Plastics	2.20	9.00	1.86	1.33
Other	4.33	2.38	4.65	2.22
Total	46.01	44.69	42.31	42.20

Table S4.1 Bill of materials of lithium ion cells (18650 form factor)

S5. Electric vehicle consumption rate

The energy consumption rates of electric vehicle models in the recent years (according to EPA tests), and assumptions documented in the literature have been listed in table S5.1. Based on these values, the consumption rate of EVs was assumed to be 250 Wh/mile, 300 Wh/mile and 350 Wh/mile for the low, baseline and high scenarios, respectively.

EV type	Vehicle consumption rate (Wh/mile)	Reference
2012 Chevrolet Volt PHEV	360	U.S. EPA and U.S. DOE (2013)
PHEV20	360	Parks et al. (2007)
Industry standard of typical EV	350	Richter et al. (2008)
PHEVs	340	Denholm and Short (2006)
2011 Nissan LEAF BEV	340	U.S. EPA and U.S. DOE (2013)
All EV types	300	Gaines and Nelson (2010)
2012 Mitsubishi i-MiEV BEV	300	U.S. EPA and U.S. DOE (2013)
PHEVs	300	Nelson and Amine (2007)
2011 Tesla Roadster BEV	300	Van Haaren (2011)
BEV, PHEV	296	Reichmuth et al. (2012)
2012 Toyota Prius PHEV	290	U.S. EPA and U.S. DOE (2013)
2006 electric vehicles	237 Wh/mile on test cycles 280 Wh/mile during use	Duvall & Knipping (2007)
BEV, PHEV	250-300	Chiang, Y. M. (2013)
PHEV	260	Marano & Rizzoni (2008)
PHEV40	250	Kang et al. (2011)
EVs in 2020 (default)	250	ANL BatPac Model (2012)
Compact EV	250	Denholm and Short (2006)

Table S5.1 EV energy consumption rate

S6. Electric vehicle LIB inflows and outflows

For the baseline scenario, the EV battery inflow (2009 to 2034) and EOL EV battery outflows (2015 to 2040) for each vehicle type *i* (BEV, PHEV10, PHEV40, HEV) using the MFA model was estimated.

S6.1 Annual inflow of LIB packs in EVs and annual outflow of LIB packs from EVs

The number of LIB pack input in EVs included batteries entering new EV as well as replacement batteries for old EVs. The battery outflows were segregated into type 1 and type 2 EOL EV batteries based on their reuse potential. As mentioned in the Chapter 2, Type 1 EOL batteries are those that have reached their end-of-lives in EV application either before or coinciding with the vehicles' end of life while Type 2 EOL batteries are obtained from EVs that reach their end-of-lives before their batteries. This modeling was conducted separately for each EV type (*i*) on an annual basis as represented for the baseline scenario in the following tables.

BEV battery MFA: Baseline Scenario															
Battery inflow						Battery lifespan (years)						Battery outflow			
Year	Entering new BEVs	Replacement in old BEVs	Total input LIBs in BEVs	Year	BEV sales	6	8	10	12			EOL LIBs from BEVs	Type 2 EOL LIBs from BEVs	Type 1 EOL LIBs from BEVs	
						EOL batteries (1st battery)				EOL second battery					
						10%	40%	40%	10%						
2009	509	0	509	2009	509										
2010	1,836	0	1,836	2010	1,836										
2011	2,667	0	2,667	2011	2,667										
2012	2,033	0	2,033	2012	2,033										
2013	2,708	0	2,708	2013	2,708										
2014	3,832	0	3,832	2014	3,832										
2015	6,183	51	6,234	2015	6,183	51						51	0	51	
2016	7,106	184	7,289	2016	7,106	184						184	0	184	
2017	8,744	470	9,214	2017	8,744	267	204					470	0	470	
2018	15,726	938	16,663	2018	15,726	203	734					938	0	938	
2019	15,448	1,338	16,785	2019	15,448	271	1,067	204	51	51	204	1,847	305	1,541	
2020	38,064	1,196	39,260	2020	38,064	383	813	734	184	184	734	3,033	1,102	1,931	
2021	51,947	1,702	53,648	2021	51,947	618	1,083	1,067	267	267	1,067	4,369	1,600	2,768	
2022	67,117	2,243	69,361	2022	67,117	711	1,533	813	203	203	813	4,276	1,220	3,057	
2023	76,965	3,348	80,313	2023	76,965	874	2,473	1,083	271	271	1,083	6,056	1,625	4,431	
2024	87,682	4,415	92,097	2024	87,682	1,573	2,842	1,533	383	383	1,533	8,247	2,299	5,948	
2025	103,465	5,042	108,507	2025	103,465	1,545	3,498	2,473	618	618	2,473	11,225	3,710	7,516	
2026	121,406	10,097	131,503	2026	121,406	3,806	6,290	2,842	711	711	2,842	17,202	4,263	12,939	
2027	138,977	11,374	150,350	2027	138,977	5,195	6,179	3,498	874	874	3,498	20,118	5,246	14,871	
2028	159,652	21,937	181,589	2028	159,652	6,712	15,226	6,290	1,573	1,573	6,290	37,663	9,435	28,228	
2029	182,757	28,475	211,233	2029	182,757	7,697	20,779	6,179	1,545	1,545	6,179	43,923	9,269	34,654	
2030	208,910	35,615	244,525	2030	208,910	8,768	26,847	15,226	3,806	3,806	15,226	73,679	22,838	50,841	
2031	237,562	41,133	278,695	2031	237,562	10,346	30,786	20,779	5,195	5,195	20,779	93,079	31,168	61,911	
2032	269,787	47,213	317,001	2032	269,787	12,141	35,073	26,847	6,712	6,712	26,847	114,331	40,270	74,060	
2033	290,204	55,284	345,488	2033	290,204	13,898	41,386	30,786	7,697	7,697	30,786	132,249	46,179	86,070	
2034	320,014	64,528	384,541	2034	320,014	15,965	48,562	35,073	8,768	8,768	35,073	152,210	52,609	99,600	
				2035		18,276	55,591	41,386	10,346	10,346	41,386	177,331	62,079	115,252	
				2036		20,891	63,861	48,562	12,141	12,141	48,562	206,158	72,844	133,314	
				2037		23,756	73,103	55,591	13,898	13,898	55,591	235,836	83,386	152,450	
				2038		26,979	83,564	63,861	15,965	15,965	63,861	270,194	95,791	174,403	
				2039		29,020	95,025	73,103	18,276	18,276	73,103	306,803	109,654	197,148	
				2040		32,001	107,915	83,564	20,891	20,891	83,564	348,826	125,346	223,480	

Table S6.1 (a) LIB inflow and outflows for BEV batteries

PHEV10 battery MFA: Baseline Scenario														
Battery inflow						Battery lifespan (years)						Battery outflow		
Year	Entering new PHEV10	Replacement in old PHEV10	Total New Batteries	Year	PHEV10 sales	6 (10%)	8 (40%)	10 (40%)	12 (10%)			EOL LIBs from PHEV10	Type 2 EOL LIBs from PHEV10	Type 1 EOL LIBs from PHEV10
						EOL batteries (1st battery)				EOL second battery				
2009	0	0	0	2009	0									
2010	0	0	0	2010	0									
2011	0	0	0	2011	0									
2012	0	0	0	2012	0									
2013	0	0	0	2013	0									
2014	23,351	0	23,351	2014	23,351									
2015	40,724	0	40,724	2015	40,724	0						0	0	0
2016	34,333	0	34,333	2016	34,333	0						0	0	0
2017	71,173	0	71,173	2017	71,173	0	0					0	0	0
2018	81,458	0	81,458	2018	81,458	0	0					0	0	0
2019	66,042	0	66,042	2019	66,042	0	0	0	0	0	0	0	0	0
2020	75,087	2,335	77,422	2020	75,087	2,335	0	0	0	0	0	2,335	0	2,335
2021	75,063	4,072	79,135	2021	75,063	4,072	0	0	0	0	0	4,072	0	4,072
2022	78,657	12,774	91,431	2022	78,657	3,433	9,341	0	0	0	0	12,774	0	12,774
2023	76,716	23,407	100,122	2023	76,716	7,117	16,290	0	0	0	0	23,407	0	23,407
2024	76,553	21,879	98,432	2024	76,553	8,146	13,733	9,341	2,335	2,335	9,341	45,230	14,011	31,220
2025	99,559	35,074	134,633	2025	99,559	6,604	28,469	16,290	4,072	4,072	16,290	75,797	24,434	51,363
2026	105,938	40,092	146,030	2026	105,938	7,509	32,583	13,733	3,433	3,433	13,733	74,425	20,600	53,825
2027	109,801	33,923	143,724	2027	109,801	7,506	26,417	28,469	7,117	7,117	28,469	105,097	42,704	62,392
2028	115,730	37,900	153,630	2028	115,730	7,866	30,035	32,583	8,146	8,146	32,583	119,359	48,875	70,484
2029	121,164	37,697	158,860	2029	121,164	7,672	30,025	26,417	6,604	6,604	26,417	103,739	39,625	64,114
2030	125,001	39,118	164,119	2030	125,001	7,655	31,463	30,035	7,509	7,509	30,035	114,205	45,052	69,153
2031	128,850	40,642	169,492	2031	128,850	9,956	30,686	30,025	7,506	7,506	30,025	115,705	45,038	70,667
2032	132,879	41,215	174,094	2032	132,879	10,594	30,621	31,463	7,866	7,866	31,463	119,872	47,194	72,678
2033	133,859	50,804	184,663	2033	133,859	10,980	39,824	30,686	7,672	7,672	30,686	127,519	46,029	81,490
2034	136,841	53,948	190,789	2034	136,841	11,573	42,375	30,621	7,655	7,655	30,621	130,501	45,932	84,569
				2035		12,116	43,920	39,824	9,956	9,956	39,824	155,596	59,736	95,860
				2036		12,500	46,292	42,375	10,594	10,594	42,375	164,730	63,563	101,167
				2037		12,885	48,466	43,920	10,980	10,980	43,920	171,151	65,881	105,271
				2038		13,288	50,000	46,292	11,573	11,573	46,292	179,018	69,438	109,580
				2039		13,386	51,540	48,466	12,116	12,116	48,466	186,090	72,698	113,391
				2040		13,684	53,151	50,000	12,500	12,500	50,000	191,837	75,001	116,836

Table S6.1 (b) LIB inflow and outflows for PHEV10 batteries

PHEV40 battery MFA: Baseline Scenario														
Battery inflow						Battery lifespan (years)						Battery outflow		
Year	Entering new PHEV40	Replacement in old PHEV40	Total New Batteries	Year	PHEV40 sales	6 (10%)	8 (40%)	10 (40%)	12 (10%)			EOL LIBs from PHEV40	Type 2 EOL LIBs from PHEV40	Type 1 EOL LIBs from PHEV40
						EOL batteries (1st battery)				EOL second battery				
2009	0	0	0	2009	0									
2010	0	0	0	2010	0									
2011	27,917	0	27,917	2011	27,917									
2012	11,560	0	11,560	2012	11,560									
2013	11,326	0	11,326	2013	11,326									
2014	32,709	0	32,709	2014	32,709									
2015	34,489	0	34,489	2015	34,489	0						0	0	0
2016	45,480	0	45,480	2016	45,480	0						0	0	0
2017	53,258	2,792	56,049	2017	53,258	2,792	0					2,792	0	2,792
2018	61,381	1,156	62,537	2018	61,381	1,156	0					1,156	0	1,156
2019	62,425	12,299	74,724	2019	62,425	1,133	11,167	0	0	0	0	12,299	0	12,299
2020	69,593	7,895	77,488	2020	69,593	3,271	4,624	0	0	0	0	7,895	0	7,895
2021	71,917	7,979	79,896	2021	71,917	3,449	4,531	11,167	2,792	2,792	11,167	35,896	16,750	19,146
2022	74,608	17,631	92,240	2022	74,608	4,548	13,083	4,624	1,156	1,156	4,624	29,192	6,936	22,256
2023	74,228	19,121	93,349	2023	74,228	5,326	13,795	4,531	1,133	1,133	4,531	30,448	6,796	23,652
2024	71,219	24,330	95,549	2024	71,219	6,138	18,192	13,083	3,271	3,271	13,083	57,039	19,625	37,414
2025	73,327	27,546	100,873	2025	73,327	6,242	21,303	13,795	3,449	3,449	13,795	62,034	20,693	41,341
2026	72,355	31,512	103,866	2026	72,355	6,959	24,552	18,192	4,548	4,548	18,192	76,992	27,288	49,704
2027	74,536	32,162	106,698	2027	74,536	7,192	24,970	21,303	5,326	5,326	21,303	85,419	31,955	53,465
2028	76,967	35,298	112,265	2028	76,967	7,461	27,837	24,552	6,138	6,138	24,552	96,679	36,828	59,850
2029	75,842	36,189	112,031	2029	75,842	7,423	28,767	24,970	6,242	6,242	24,970	98,614	37,455	61,159
2030	74,956	36,965	111,921	2030	74,956	7,122	29,843	27,837	6,959	6,959	27,837	106,558	41,756	64,802
2031	75,799	37,024	112,823	2031	75,799	7,333	29,691	28,767	7,192	7,192	28,767	108,940	43,150	65,791
2032	76,778	35,723	112,501	2032	76,778	7,235	28,488	29,843	7,461	7,461	29,843	110,331	44,765	65,566
2033	79,098	36,785	115,882	2033	79,098	7,454	29,331	29,691	7,423	7,423	29,691	111,012	44,537	66,476
2034	78,193	36,639	114,831	2034	78,193	7,697	28,942	28,488	7,122	7,122	28,488	107,858	42,731	65,126
				2035		7,584	29,815	29,331	7,333	7,333	29,331	110,726	43,996	66,730
				2036		7,496	30,787	28,942	7,235	7,235	28,942	110,637	43,413	67,224
				2037		7,580	30,337	29,815	7,454	7,454	29,815	112,453	44,722	67,731
				2038		7,678	29,982	30,787	7,697	7,697	30,787	114,627	46,180	68,447
				2039		7,910	30,320	30,337	7,584	7,584	30,337	114,071	45,505	68,566
				2040		7,819	30,711	29,982	7,496	7,496	29,982	113,486	44,973	68,513

Table S6.1 (c) LIB inflow and outflows for PHEV40 batteries

HEV Battery MFA: Baseline Scenario																
Battery inflow								Battery lifespan (years)						Battery outflow		
Year	Entering new HEV	Replacement in old HEV	Total New Batteries	Year	HEV sales	% HEVs Using LIBs	HEV sold using LIBs	6 (10%)	8 (40%)	10 (40%)	12 (10%)			EOL LIBs from HEV	Type 2 EOL LIBs from HEV	Type 1 EOL LIBs from HEV
								EOL batteries (1st battery)				EOL second battery				
2009	0	0	0	2009	304,495	0%	0									
2010	11,570	0	11,570	2010	283,456	4%	11,570									
2011	33,046	0	33,046	2011	285,397	12%	33,046									
2012	56,928	0	56,928	2012	294,128	19%	56,928									
2013	113,999	0	113,999	2013	334,643	34%	113,999									
2014	174,162	0	174,162	2014	397,448	44%	174,162									
2015	226,495	0	226,495	2015	432,858	52%	226,495	0						0	0	0
2016	276,332	1,157	277,489	2016	460,554	60%	276,332	1,157						1,157	0	1,157
2017	318,729	3,305	322,033	2017	490,352	65%	318,729	3,305	0					3,305	0	3,305
2018	358,120	10,321	368,440	2018	511,599	70%	358,120	5,693	4,628					10,321	0	10,321
2019	391,705	24,618	416,323	2019	532,127	74%	391,705	11,400	13,218	0	0	0	0	24,618	0	24,618
2020	415,040	40,187	455,227	2020	553,387	75%	415,040	17,416	22,771	4,628	1,157	1,157	4,628	51,757	6,942	44,815
2021	465,773	68,249	534,022	2021	582,216	80%	465,773	22,650	45,600	13,218	3,305	3,305	13,218	101,295	19,828	81,468
2022	540,504	97,298	637,803	2022	635,888	85%	540,504	27,633	69,665	22,771	5,693	5,693	22,771	154,226	34,157	120,069
2023	616,741	122,471	739,212	2023	685,268	90%	616,741	31,873	90,598	45,600	11,400	11,400	45,600	236,470	68,400	168,071
2024	690,749	146,345	837,094	2024	727,105	95%	690,749	35,812	110,533	69,665	17,416	17,416	69,665	320,507	104,497	216,010
2025	749,102	166,662	915,764	2025	749,102	100%	749,102	39,170	127,491	90,598	22,650	22,650	90,598	393,157	135,897	257,260
2026	770,004	184,752	954,755	2026	770,004	100%	770,004	41,504	143,248	110,533	27,633	27,633	110,533	461,084	165,799	295,285
2027	786,097	203,259	989,356	2027	786,097	100%	786,097	46,577	156,682	127,491	31,873	31,873	127,491	521,988	191,237	330,751
2028	798,356	220,066	1,018,423	2028	798,356	100%	798,356	54,050	166,016	143,248	35,812	35,812	143,248	578,186	214,872	363,314
2029	805,968	247,983	1,053,951	2029	805,968	100%	805,968	61,674	186,309	156,682	39,170	39,170	156,682	639,688	235,023	404,665
2030	815,691	285,277	1,100,968	2030	815,691	100%	815,691	69,075	216,202	166,016	41,504	41,504	166,016	700,317	249,024	451,293
2031	823,508	321,607	1,145,115	2031	823,508	100%	823,508	74,910	246,696	186,309	46,577	46,577	186,309	787,379	279,464	507,916
2032	833,867	353,300	1,187,167	2032	833,867	100%	833,867	77,000	276,300	216,202	54,050	54,050	216,202	893,805	324,303	569,502
2033	840,145	378,250	1,218,396	2033	840,145	100%	840,145	78,610	299,641	246,696	61,674	61,674	246,696	994,991	370,045	624,947
2034	846,415	387,837	1,234,252	2034	846,415	100%	846,415	79,836	308,001	276,300	69,075	69,075	276,300	1,078,586	414,450	664,137
				2035				80,597	314,439	299,641	74,910	74,910	299,641	1,144,137	449,461	694,676
				2036				81,569	319,342	308,001	77,000	77,000	308,001	1,170,915	462,002	708,913
				2037				82,351	322,387	314,439	78,610	78,610	314,439	1,190,835	471,658	719,177
				2038				83,387	326,276	319,342	79,836	79,836	319,342	1,208,019	479,014	729,006
				2039				84,015	329,403	322,387	80,597	80,597	322,387	1,219,385	483,581	735,805
				2040				84,641	333,547	326,276	81,569	81,569	326,276	1,233,879	489,414	744,465

Table S6.1 (d) LIB inflow and outflows for HEV batteries

S6.2 Calculation of materials mass of annual EV LIB inflows and outflows

In the MFA model it was assumed that the percentage of LIBs of a given chemistry in waste stream is same as the percentage of new LIBs of that chemistry entering EV use. Hence, using m_j, the mass of a cylindrical cell of chemistry *j*, along with the percentage of LIBs of cathode chemistry *j* in EV battery waste stream (PC_j) and the number of cells per battery pack for EV type *i* and cathode chemistry *j* ($D_{i,j}$), the material mass of EV battery inflows into new EVs ($MI_{s,t}$) as well as the material mass of replacement batteries in old EVs ($MI_{w,t}$) for given year *"t"* was estimated as,

$$MI_{s,t} = \sum_i \sum_j (S_{i,t} * PC_j * D_{i,j} * m_j)$$

$$MI_{w,t} = \sum_i \sum_j (W_{i,t} * PC_j * D_{i,j} * m_j)$$

Where,

$S_{i,t}$ = Sales of new EVs of type *i* that use lithium ion batteries in year *t*
$W_{i,t}$ = Non-EOL EVs of type *i* requiring a replacement LIB in year *t*

The following table represents the LIB inflow into new EVs and as replacement in old EVs on an annual basis (2009-2034) for the baseline scenario. These results are represented in a cumulative format in the Sankey diagram (Figure 2.6 in the Chapter 2). The annual LIB inflow is calculated from tables S6.1 (a) through S6.1 (d).

	LIB inflow into new EVs		Replacement LIBs in old EVs	
Year	Number of LIB packs	Cell mass (metric tons)	Number of LIB packs	Cell mass (metric tons)
2009	509	124	0	0
2010	13,406	829	0	0
2011	63,630	4,819	0	0
2012	70,521	3,652	0	0
2013	128,034	5,678	0	0
2014	234,055	10,942	0	0
2015	307,891	13,919	51	12
2016	363,252	16,828	1,341	83
2017	451,904	20,501	6,567	531
2018	516,684	24,681	12,414	697
2019	535,619	25,414	38,255	2,495
2020	597,784	32,725	51,614	2,555
2021	664,699	38,035	82,003	3,663
2022	760,887	44,592	129,947	6,059
2023	844,650	49,413	168,347	7,618
2024	926,203	54,130	196,969	9,199
2025	1,025,453	60,765	234,323	10,742
2026	1,069,702	65,888	266,452	13,145
2027	1,109,411	71,040	280,718	13,969
2028	1,150,705	76,905	315,202	17,549
2029	1,185,730	82,801	350,345	20,155
2030	1,224,558	89,491	396,975	23,250
2031	1,265,719	96,916	440,405	25,841
2032	1,313,311	105,315	477,451	28,241
2033	1,343,307	110,770	521,122	31,410
2034	1,381,462	118,209	542,951	34,046
Cumulative (2009-2034)	18,549,085	1,224,380	4,513,453	251,261

Table S6.2 (a) Estimation of material mass of EV battery inflows (Baseline scenario)

Similarly, for any year *t*, the material outflows (*MO*) from EOL EV batteries of a given type "*g*" (type 1 or type 2) was estimated using the same variables m_j, PC_j and $D_{i,j}$ along with the number of LIB packs in EV battery waste stream (B_t) and the percentage of type 1 or type 2 EOL EV batteries generated from EV type *i* in that year ($PE_{g,i,t}$).

$$MO_{g,t} = B_t * \sum_i \sum_j (PE_{g,i,t} * PC_j * D_{i,j} * m_j)$$

The following table represents the EOL battery outflows on an annual basis from 2015 to 2040 and classifies them into type 1 and type 2 EOL batteries based on tables S6.1 (a) through S6.1 (d). These results are represented in a cumulative format in the Sankey diagram (Figure 2.6 in Chapter 2).

	Type 1 EOL LIB packs		**Type 2 EOL LIB packs**	
Year	Number of LIB packs	Cell mass (metric tons)	Number of LIB packs	Cell mass (metric tons)
2015	51	12	0	0
2016	1,341	83	0	0
2017	6,567	531	0	0
2018	12,414	697	0	0
2019	38,459	2,545	305	74
2020	56,976	2,886	8,044	498
2021	107,455	5,591	38,178	2,892
2022	158,155	7,520	42,313	2,191
2023	219,560	9,889	76,820	3,407
2024	290,591	13,576	140,433	6,565
2025	357,480	16,309	184,735	8,352
2026	411,753	19,876	217,951	10,097
2027	461,479	22,169	271,142	12,301
2028	521,876	27,422	310,010	14,808
2029	564,592	30,321	321,372	15,248
2030	636,089	36,340	358,670	19,635
2031	706,285	41,055	398,819	22,821
2032	781,806	46,078	456,532	26,755
2033	858,982	51,175	506,790	29,648
2034	913,432	55,698	555,722	32,478
2035	972,519	61,002	615,272	36,459
2036	1,010,618	66,066	641,821	39,533
2037	1,044,628	71,228	665,646	42,624
2038	1,081,436	77,090	690,423	46,143
2039	1,114,911	82,964	711,438	49,681
2040	1,153,294	89,743	734,735	53,695
Cumulative (2015-2040)	13,482,749	837,866	7,947,172	475,902

S6.2 (b) Estimation of material mass of EV battery outflows for type 1 and type 2 EOL EV batteries (Baseline scenario)

S6.3 Scenario specific annual estimates of EV LIB material outflows

EV LIB material outflows (Metric tons)			
Year	**Low Scenario**	**Baseline Scenario**	**High Scenario**
2015	8	12	702
2016	33	83	1,858
2017	208	531	4,195
2018	245	697	8,421
2019	1,083	2,619	13,843
2020	1,205	3,384	22,824
2021	4,005	8,482	35,847
2022	2,587	9,711	54,998
2023	3,085	13,296	70,882
2024	4,518	20,141	98,071
2025	6,309	24,661	113,346
2026	7,283	29,973	135,639
2027	8,895	34,471	149,798
2028	10,770	42,230	180,872
2029	11,495	45,570	173,598
2030	13,976	55,976	193,212
2031	15,007	63,877	205,598
2032	16,800	72,834	219,715
2033	18,563	80,824	231,105
2034	20,433	88,177	242,448
2035	23,060	97,462	261,704
2036	25,636	105,600	277,471
2037	28,692	113,854	299,258
2038	31,992	123,234	315,595
2039	35,344	132,647	336,585
2040	38,285	143,440	343,929

Table S6.3 Scenario specific annual estimates of EV LIB material outflows

S7. Uncertainty analysis of MFA parameters

Recognizing the uncertainty associated with EV lifespan, this MFA model assumed a moderate EV lifespan of 10 years, hence, a sensitivity analysis was conducted assuming a 16 year EV lifespan, this being the higher end lifespan assumed for conventional vehicle studies and the current U.S. DOE estimate of the lifespan of a typical light duty vehicle in the United States (Davis et al., 2013). Moreover, the wide disparity observed between the low and high scenario in our analysis is indicative of the uncertainty in the EV sales, their battery lifespan as well as the parameters determining the number of cells per EV battery pack. The impact of these uncertainties on the MFA model is analyzed in the subsequent sections.

S7.1 Impact of EV sales and battery lifespan distribution on MFA results

The baseline EV sales projections were combined with the entire range of modeled battery lifespan distributions to analyze the impact of battery lifespan on waste EV LIB outflows. Similarly, the baseline EV LIB lifespan distribution was combined with the high and low scenario EV sales figures. The EOL EV battery outflows in each of these scenarios were compared with the battery outflows in the initial baseline scenario (i.e. baseline EV sales and baseline LIB lifespan distribution). These results are shown below:

Year	EOL EV LIB pack units (Baseline Sales-Baseline Scenario Lifespan Distribution)	EOL EV LIB pack units (Baseline Sales-Low Scenario Lifespan Distribution)	EOL EV LIB pack units (Baseline Sales-High Scenario Lifespan Distribution)	EOL EV LIB pack units (High Sales- Baseline Scenario Lifespan Distribution	EOL EV LIB pack units (Low Sales- Baseline Scenario Lifespan Distribution
2015	51	25	6,490	50	355
2016	1,341	670	10,404	1,337	1,692
2017	6,567	3,309	28,869	6,565	7,237
2018	12,414	6,878	45,192	20,298	11,167
2019	38,764	22,818	83,032	52,755	32,233
2020	65,020	42,739	130,106	126,874	43,824
2021	145,633	111,033	225,484	246,630	92,728
2022	200,468	147,197	285,559	448,144	96,205
2023	296,381	227,602	390,018	670,450	126,455
2024	431,024	350,702	535,612	983,810	177,476
2025	542,214	447,648	648,356	1,262,360	238,619
2026	629,704	522,312	748,959	1,398,700	278,716
2027	732,622	619,044	868,586	1,541,070	347,008
2028	831,886	704,174	984,839	1,701,250	405,137
2029	885,964	744,027	1,055,384	1,683,420	436,642
2030	994,759	834,316	1,172,180	1,798,170	498,093
2031	1,105,104	927,134	1,293,845	1,876,030	531,040
2032	1,238,339	1,045,923	1,430,506	1,990,030	582,229
2033	1,365,772	1,156,484	1,558,466	2,084,930	631,460
2034	1,469,154	1,251,164	1,667,943	2,161,360	676,447
2035	1,587,790	1,362,092	1,792,375	2,271,950	732,520
2036	1,652,439	1,418,606	1,863,891	2,351,960	776,866
2037	1,710,275	1,469,129	1,927,747	2,440,890	829,062
2038	1,771,859	1,522,510	1,996,792	2,488,430	878,135
2039	1,826,349	1,569,325	2,054,594	2,557,040	942,932
2040	1,888,028	1,621,959	2,125,742	2,554,530	985,513
Cumulative (2015 -2040)	21,429,921	18,128,819	24,930,969	34,719,032	10,359,792

Table S7.1 Impact of EV sales and LIB lifespan distribution on EV battery MFA results

S7.2 Impact of battery pack energy storage on MFA results

Sensitivity analysis was conducted wherein the EOL EV battery pack numbers for the baseline scenario were used along with the EV battery pack energy storage (determining the number of cells per battery pack) for the low as well as the high scenario to determine the material mass of the EV LIB waste stream. These material masses were then compared with material outflows in the initial baseline scenario as shown below:

Year	EV LIB material outflows (Metric Tonnes): Baseline scenario cells per battery pack assumption	EV LIB material outflows (Metric Tonnes): Low scenario cells per battery pack assumption	EV LIB material outflows (Metric Tonnes): High scenario cells per battery pack assumption
2015	12	9	16
2016	83	58	118
2017	531	385	726
2018	697	489	995
2019	2,619	1,874	3,641
2020	3,384	2,368	4,848
2021	8,482	6,008	11,954
2022	9,711	6,774	13,967
2023	13,296	9,205	19,306
2024	20,141	14,046	28,974
2025	24,661	17,198	35,478
2026	29,973	20,953	42,988
2027	34,471	24,120	49,377
2028	42,230	29,734	60,011
2029	45,570	32,046	64,862
2030	55,976	39,623	78,997
2031	63,877	45,260	90,038
2032	72,834	51,628	102,612
2033	80,824	57,290	113,877
2034	88,177	62,532	124,166
2035	97,462	69,259	136,866
2036	105,600	75,248	147,755
2037	113,854	81,342	158,744
2038	123,234	88,284	171,193
2039	132,647	95,268	183,635
2040	143,440	103,273	197,913
Cumulative (2015-2040)	1,313,787	934,277	1,843,057

Table S7.2 Mass of lithium ion cells in EV battery waste stream under baseline scenario EOL battery pack numbers and different assumptions of cells per battery pack.

S7.3 Impact of extended EV lifespan on MFA results

When the baseline scenario was remodeled with an extended EV lifespan of 16 years, only a small difference was observed in the number of EOL EV LIBs entering the waste streams when compared with the initial baseline scenario results that assumed a 10 year EV lifespan. While in case of 10 year EV lifespan, Type 2 EOL batteries would start entering the waste stream a few years early (i.e. year 2019) owing to shorter EV lifespan, they would enter the waste stream as late as year 2025 when the EV lifespan is extended to 16 years. In the initial baseline scenario with the 10 year EV lifespan, there was scope for only a single battery replacement during the EV lifetime. However, when the EV lifespan was extended, a third LIB pack was needed by few vehicles which were 12 years or older. Acknowledging that this may not be feasible from a cost perspective, a variant to the 16 year EV lifespan-baseline scenario MFA modeling was introduced, wherein the number of replacement LIB pack was restricted to one during the lifetime of the EV (assuming that the vehicle would become obsolete before 16 years, if its second battery reached EOL). Apart from annual differences in EV LIB outflows, there was not much difference observed in the cumulative (2015 to 2040) outflows of these batteries into the waste stream among these different scenarios of EV lifespan as indicated in table S7.3.1.

Year	EOL EV LIB pack units (10 year EV lifespan)	EOL EV LIB pack units (16 year EV lifespan	EOL EV LIB pack units (16 year EV lifespan with restriction of one replacement battery)
2015	51	51	51
2016	1,341	1,341	1,341
2017	6,567	6,567	6,567
2018	12,414	12,414	12,414
2019	38,764	38,459	38,459
2020	65,020	56,976	56,976
2021	145,633	107,511	107,511
2022	200,468	159,630	159,630
2023	296,381	226,600	226,600
2024	431,024	299,421	299,421
2025	542,214	377,163	377,117
2026	629,704	456,546	455,340
2027	732,622	569,220	563,493
2028	831,886	651,079	644,732
2029	885,964	766,967	755,444
2030	994,759	956,039	934,974
2031	1,105,104	1,109,246	1,081,536
2032	1,238,339	1,252,149	1,219,457
2033	1,365,772	1,426,853	1,386,181
2034	1,469,154	1,561,637	1,515,135
2035	1,587,790	1,654,225	1,606,020
2036	1,652,439	1,771,156	1,717,355
2037	1,710,275	1,889,699	1,829,876
2038	1,771,859	2,034,087	1,965,607
2039	1,826,349	2,163,632	2,087,613
2040	1,888,028	2,291,650	2,208,292
Cumulative EOL EV LIB outflows (2015 to 2040)	21,429,921	21,840,317	21,257,142

Table S7.3.1 EOL lithium ion battery outflows under different EV lifespan scenarios

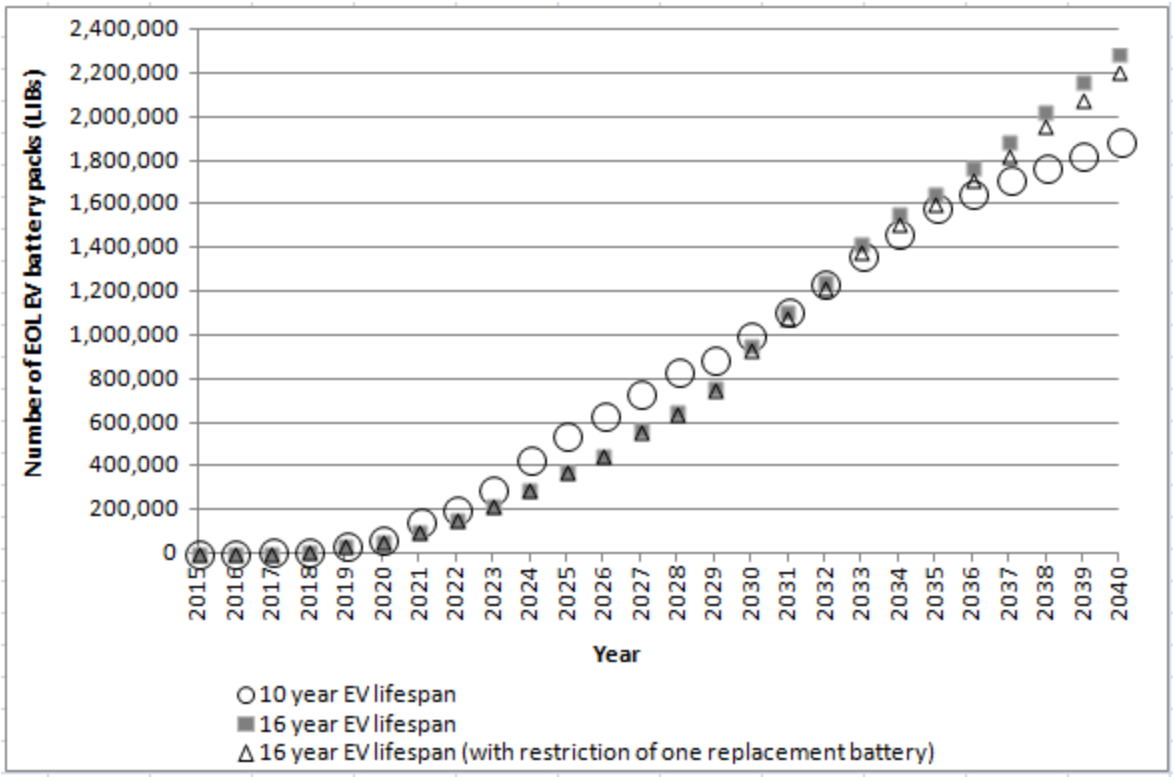


Figure S7.3.1 EOL lithium ion battery outflows under different EV lifespan scenarios

As a result, the baseline scenario material outflow of lithium ion cells in the EV battery waste stream was nearly 1.3 million metric tons in all the three EV lifespan variants on a cumulative basis between 2015 and 2040. Although, differences were observed in the waste EV LIB material outflows in individual years during the timeline of this analysis due to extended EV lifespan assumption or assuming single battery replacement over the extended EV lifespan as shown in figure S7.3.2.

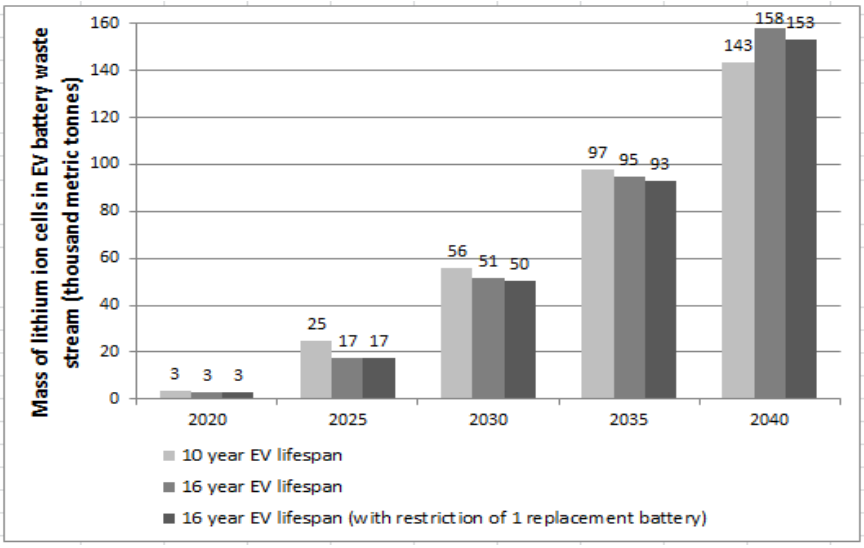


Figure S7.3.2 Mass of Li-Ion cells in EV battery waste stream under different EV lifespan scenarios

In terms of type 2 EOL EV batteries, a reduction was observed in the percentage of these type of batteries accrued in the waste stream between 2015 and 2040 due to extended EV lifespan.

EV lifespan in baseline scenario	Percentage of Type 1 EOL batteries	Percentage of Type 2 EOL batteries
10 years	63%	37%
16 years	77%	23%
16 years (with restriction of 1 replacement battery)	80%	20%

Table S7.3.2. Percentage of type 1 and type 2 EOL EV batteries accrued in the waste stream between 2015 and 2040 under different EV lifespan scenarios

S8. Prismatic lithium ion cells

S8.1 Bill of materials of prismatic cells

The material composition of prismatic lithium ion cells was estimated using the Argonne National Laboratory BatPac simulation model (ANL, 2012). For any given cathode chemistry, the BatPac model provides cell design estimates for 7 cells based on their capacity and size. An intermediate size (i.e. battery 4) cell design was used for our model. The estimation of the bill of materials of a $LiFePO_4$ prismatic cell is explained as follows:

Data obtained from ANL BatPaC model:

Prismatic Cell (LiFePO4)	**Mass (g)**
Active materials (Positive electrode)	214.22
Active materials (Negative electrode)	119.57
Carbon	14.44
Binder	18.33
Al (Positive foil)	43.40
Cu (Negative foil)	92.56
Separator (PP, EP/PP)	14.88
Electrolyte	155.74
Positive terminal assembly (Aluminum)	7.52
Negative terminal assembly (nickel plated copper sheet)	24.84
Cell container (PET-AL-PP)	33.91
Total (per cell)	739.39

Table S8. 1.1 Mass of prismatic li-ion cell ($LiFePO_4$) components obtained from ANL BatPac model (2012)

The BatPac model gives only the integrated mass of cathode and anode active materials and does not categorize them into the constituent materials. The anode is typically all graphitic so all the active negative electrode should be graphite. To calculate the mass percent of each element in the cathode, the molar masses of each element (found in periodic table) was used. For instance, if the cathode chemistry is LiFePO4, it implies that for one mole of Li, Fe, and P, it contains 4 moles of oxygen. Therefore,

Li - 1 mole = 6.94g
Fe - 1 mole = 55.845g
P - 1mole = 30.974g
O - 1mole - 15.99g * 4 moles = 63.96g

Total molar mass = 157.719g

So the mass percentage of each element in the cathode structure would be:

Li - 6.94g/157.719g = 0.044 or 4.4%
Fe - 55.845g/157.719g = 0.354 or 35.4%
P - 30.974g/157.719g = 0.196 or 19.6%
O - 63.96g/157.719g = 0.406 or 40.6%

Therefore for a prismatic cell which has 214.22 g of active material ($LiFePO_4$) the elemental masses would be:

Li = 0.044 * 214.22g = 9.43g
Fe = 0.354 * 214.22g = 75.85g
P = 0.196 * 214.22g = 42.07g
O = 0.406*214.22g = 86.87g

Total aluminum present in the prismatic cell was calculated as the sum of aluminum present in positive foil, positive terminal assembly and the cell container. Aluminum present in the $LiFePO_4$ cell container was estimated as a product of cell width, cell length, thickness of cell container aluminum layer, and density of aluminum from the following data from BatPac model:

Width of cell (mm) = 117 (ANL, 2012)
Length of cell (mm) = 375 (ANL, 2012)
Thickness of cell container aluminum layer (μm) = 100 (ANL, 2012)
Density of aluminum (g/cm^3) = 2.7
Hence, aluminum in cell container (g) = 11.87
Plastic in cell container (g) = 33.91-11.87 = 22.04

Plastic material present in the prismatic cell was estimated as the amount of plastics present in separator and the cell container. In a similar way, the bill of materials of prismatic cells of $LiMn_2O_4$ and NCM chemistry was estimated as shown in table S8.1.2. For the NCM chemistry,

LIB type NCM441 (Li1.05(Ni4/9Mn4/9Co1/9)0.95O2-Graphite) was used in our model. NCM-441 is less expensive than the NCM-333 (Li1.05(Ni1/3Mn1/3Co1/3)0.95O2-Graphite) due to significantly less amount of cobalt present (ANL, 2012) and hence, would be likely to be used in EV application in the future.

Cathode Chemistry	**LiMn2O4**	**LiFePO4**	**NCM**
Material, *y* (grams)			
Aluminum	50.00	62.78	45.08
Cobalt	0	0	10.72
Copper	94.40	117.40	86.62
Lithium	10.20	9.43	12.56
Manganese	161.54	0	39.97
Nickel	0	0	42.70
Steel	0	0	0
Iron	0	75.85	0.00
Graphite	99.17	119.57	109.90
Carbon	17.92	14.44	10.86
Binder	20.15	18.33	14.83
Plastic	29.74	36.92	25.88
Electrolyte (LiPF6 in EC)	97.29	155.74	78.33
Other	94.03	128.94	55.11
Total	674.45	739.39	532.56

Table S8.1.2 Bill of materials of lithium ion cells (prismatic form factor) as estimated from ANL BatPac model (2012)

S8.2 Number of prismatic cells per EV battery pack

The number of cells per LIB pack for a given EV type and a given battery chemistry ($D_{i,j}$) was estimated as follows:

$$D_{i,j} = (Epack_i(Wh) / Ecell_j(Wh))$$

The cell energy of prismatic cells was estimated as the product of cell capacity and average voltage as obtained from the BatPac model.

Cathode chemistry	**Cell Capacity (Ah)**	**Average voltage (V)**	**Cell Energy (Wh)**
LMO	26.58	3.95	105.09
LFP	32.13	3.28	105.46
NCM	28.19	3.75	105.70

Table 8.2.1 Cell energy estimation of prismatic lithium ion cells

Baseline scenario parameters were used to estimate the LIB battery energy storage for BEV (39 kWh), PHEV10 (4.4 kWh), PHEV40 (18 kWh) and HEV (5.3 kWh), as indicated in table 2.1 of Chapter 2. Based on the cell and battery pack energy storage, the number of prismatic lithium ion cells per EV battery pack was estimated as follows:

Cathode chemistry	No. of cells per BEV battery pack	No. of cells per PHEV10 battery pack	No. of cells per PHEV40 battery pack	No. of cells per HEV battery pack
LMO	373	42	169	51
LFP	372	42	169	51
NCM	371	42	168	50

Table S8.2.2 Number of cells per battery pack (prismatic LIBs)

S8.3 Composition of EV battery waste stream in prismatic cells scenario

The composition of EV battery waste stream when all LIB packs consisted of cylindrical cells (Base Case) and 90% of the battery packs consisted of prismatic cells is shown in table S8.3. The chemistry mix of EV battery waste stream was assumed to be same in both scenarios (10% LCO, 30% each of LMO, LFP and NCM cells based LIBs). In the 90% prismatic scenario only the LCO cells were of 18650 form factor (consistent with adoption by some auto makers, like Tesla) while the remaining 90% of the LIBs in the EV battery waste stream consisted of prismatic cells.

	Base Case Scenario	90% Prismatic Scenario
Aluminum	4.34%	7.83%
Cobalt	3.97%	2.15%
Copper	5.71%	14.56%
Lithium	1.38%	1.69%
Manganese	8.81%	9.38%
Nickel	2.44%	2.09%
Steel	22.45%	1.57%
Iron	3.29%	3.52%
Carbon	24.47%	20.06%
Binder, plastic	10.37%	7.46%
Electrolyte materials (lithium salt in organic carbonate)	5.15%	15.84%
Others	7.62%	13.82%
Total	100.00%	100.00%

Table S8.3 Composition of EV battery waste stream

S9. Commodity values

Prices of LIB materials were obtained from United States Geological Survey data (USGS, 2012), London Metal Exchange (2012) and Shanghai Metals Market (2012).

Lithium ion battery material	Material price
	$/g
Cobalt	0.03648
Manganese	0.00357
Copper	0.00956
Aluminum	0.00264
Steel	0.00064
Iron	0.00064
Nickel	0.02423
Lithium	0.06220

Table S9.1 Commodity values of LIB material

S10. Recycling efficiency of LIB materials

Metal in LIB	Recycling Efficiency (%)	Literature source
Cobalt	89	Li et al. (2009)
Nickel	62	Graedel et al. (2011)
Iron/Steel	52	Fenton (2003)
Aluminum	42	Plunkert (2005)
Copper	90	Ruhrberg (2006)

Table S10.1 Recycling efficiency of LIB materials

S11. USGS definitions (USGS, 2012)

Reserves: That part of the reserve base which could be economically extracted or produced at the time of determination. The term reserves need not signify that extraction facilities are in place and operative. Reserves include only recoverable materials; thus, terms such as “extractable reserves” and “recoverable reserves” are redundant and are not a part of this classification system.

Reserve Base: That part of an identified resource that meets specified minimum physical and chemical criteria related to current mining and production practices, including those for grade, quality, thickness, and depth. The reserve base is the in-place demonstrated (measured plus indicated) resource from which reserves are estimated. It may encompass those parts of the resources that have a reasonable potential for becoming economically available within planning horizons beyond those that assume proven technology and current economics. The reserve base includes those resources that are currently economic (reserves), marginally economic (marginal reserves), and some of those that are currently sub-economic (sub-economic resources).

APPENDIX B

S.1. EV lithium-ion battery production

For both battery systems (lithium-ion and lead-acid) material production, processing and use phase stages were modeled in SimaPro LCA software using the ecoinvent database version 2.2 (ecoinvent Centre, 2010). U.S. based electricity grid mix was used for electricity input data. For metal inputs, instead of assuming 100% primary metal usage a production mix of the metals was considered. Steel components were modeled to be composed of low alloyed steel which has about 37% recycled material content (ecoinvent Centre, 2010). Aluminum components were assumed to be composed of a production mix of 68% primary and 32% secondary aluminum (ecoinvent Centre, 2010). Copper components were modeled as comprising of 85% primary and 15% secondary copper based on average copper consumption mix (International Institute for Sustainable Development, 2010).

The lithium-ion battery (LIB) production stage includes the manufacture of various battery parts such as the lithium manganese oxide ($LiMn_2O_4$) cathode, graphite anode, separator composed of plastic material, electrolyte salt (Lithium hexafluorophosphate [$LiPF_6$]), electrolyte solvent (Ethylene Carbonate), battery pack components as well as cell assembly and battery pack assembly.

The EV LIB pack data has been obtained from the Argonne National Laboratory BatPac Model (ANL, 2011) by running the model for a 24 kWh BEV battery consisting of 48 modules, with 4 cells per module (2 in parallel and 2 in series). The battery pack comprised of 192 prismatic cells. The BatPac model provided a reasonable estimation of material inputs for LIB cells, modules and pack. Remaining data on battery management system (BMS) as well as LIB manufacturing processes for various components has been collected from literature sources and past LCA studies, stoichiometry, material properties and ecoinvent database.

S.1.1. Lithium-ion cell

Bill of materials for lithium manganese oxide cell was calculated from the outputs of Argonne National Laboratory BatPac model (ANL, 2011). Table 1 shows the composition of a single LIB cell as obtained from the model for a LIB pack parameterized based on specific inputs (pack energy, cells per pack, etc.)

Li-ion ($LiMn_2O_4$) cell component	**Weight (g)**
Cathode	402.14
Anode	213.26
Positive terminal assembly	7.31
Negative terminal assembly	24.14
Separator	13.9
Electrolyte	117.52
Cell container	41.6
Total cell mass	**819.85**

Table 1. LIB cell composition

1.1 Cathode

Description	Input	Output	Unit	ecoinvent material/process	Reference/Remarks
Functional unit					
LIB cathode		402.14	g		ANL (2011)
Materials					
Cathode active material (LiMn2O4)	321.96		g	Lithium manganese oxide, at plant/GLO U	ANL (2011)
Carbon (as conductor)	21.71		g	Carbon black, at plant/GLO U	ANL (2011)
Binder (PVDF)	18.09		g	Polyvinylfluoride, at plant/US U	ANL (2011); PVF used as a proxy for PVDF
N-Methylpyrrolidone NMP (binder solvent)	177.08		g	N-methyl-2-pyrrolidone, at plant/RER U	Based on active material and NMP ratio in cathode from Majima (2001)
Aluminum foil	40.39		g	Aluminium, production mix, at plant/RER U	ANL (2011)
Energy and processes					
Aluminum foil production	40.39		g	Sheet rolling, aluminium/RER U	ecoinvent standard process
Electricity	0.0008043		KWh	Electricity, medium voltage, at grid/US U	Calculated from Notter et al. (2010)
Process heat	0.26		MJ	Heat, natural gas, at industrial furnace >100kW/RER U	Based on specific heat of all materials, heat of vaporization of NMP and a heating efficiency of 60%.
Transport					
Rail Transport	0.324		tkm	Transport, freight, rail/RER U	ecoinvent standard distances
Road Transport	0.058		tkm	Transport, lorry >16t, fleet average/RER U	ecoinvent standard distances
Infrastructure					
Chemical plant/organic	1.6E-10		p	Chemical plant, organics/RER/I U	ecoinvent Centre, 2010

Table 2 Input-output table for the production of LIB cathode

LIB cathode production requires the mixing of $LiMn_2O_4$ active material, binder, carbon black and the solvent into a slurry which is then subsequently used to coat the current collector made of aluminum foil. This process requires the heating of the slurry to 120 degree Celsius as well as thermal energy to evaporate any water and dry the cathode (Saevarsdottir, 2011).

The solvent for the binder used in both cathode and anode production is N-Methylpyrrolidone (NMP). Since, the electrode paste containing this solvent is heated to dry the electrode, the NMP vapor if released into the environment could be an environmental concern. Also high cumulative energy demand of NMP (ecoinvent Centre, 2010) makes it an expensive solvent. Hence, both

from environmental and economic point of view, battery manufacturers aim to recover NMP. A 98% recovery rate of NMP has been assumed based on U.S. Department of Energy [U.S.DOE] (2010). LCI data for NMP recovery was based on generic inventory data for solvent recovery from Geisler et al. (2004).

NMP Recovery

Description	Input	Output	Unit	ecoinvent material/process	Reference/Remarks
Avoided product					
NMP recovered		173.535	g	N-methyl-2-pyrrolidone, at plant/RER U	98% recovery based on LG Chem Ltd. and Compact Power, Inc. (U.S. DOE, 2010)
Others					
Water for NMP recovery	14166		g	Water, decarbonised, at plant, RER U	Geisler et al., 2004
Nitrogen for NMP recovery	2.5		g	Nitrogen, via cryogenic air separation, production mix, at plant, gaseous EU-27 S	Geisler et al., 2004
Steam for NMP recovery	260.00		g	Steam, for chemical processes, at plant/RER U	Geisler et al., 2004
Energy and processes					
Electricity for NMP recovery	0.0087		KWh	Electricity, medium voltage, at grid/US U	Geisler et al., 2004

Table 3 Input-output table for NMP recovery post-cathode production

1.2 Anode

Description	Input	Output	Unit	ecoinvent material/process	Reference/Remarks
Functional unit					
LIB cathode		213.26	g		ANL (2011)
Materials					
Anode active material (graphite)	119.47		g	Graphite, battery grade, at plant/CN U	ANL (2011)
Binder (PVDF)	6.29		g	Polyvinylfluoride, at plant/US U	ANL (2011)
NMP (binder solvent)	119.47		g	N-methyl-2-pyrrolidone, at plant/RER U	Based on active material and NMP ratio from Majima (2001)
Copper foil	87.51		g	85% Copper, primary, at refinery/RER U; 15% Copper, primary, at refinery/RER U	ANL (2011)
Energy and processes					
Copper foil production	87.51		g	Sheet rolling, copper/RER U	ecoinvent standard process
Electricity	0.00043		KWh	Electricity, medium voltage, at grid/US U	Notter et al. (2010)
Process heat	0.16		MJ	Heat, natural gas, at industrial furnace >100kW/RER U	Based on specific heat of materials, heat of vaporization of NMP and a heating efficiency of 60%
Transport					
Rail Transport	0.162		tkm	Transport, freight, rail/RER U	ecoinvent standard distances
Road Transport	0.033		tkm	Transport, lorry >16t, fleet average/RER U	ecoinvent standard distances
Infrastructure					
Chemical plant/organic	8.5E-10		p	Chemical plant, organics/RER/I U	ecoinvent Centre, 2010

Table 4 Input-output table for the production of LIB anode

LIB anode production requires the mixing of graphite, binder, and the solvent into a slurry which is then subsequently used to coat the current collector made of copper foil. This process requires heating of the slurry to 120 degree Celsius as well as thermal energy to evaporate any water and dry the anode (Saevarsdottir, 2011; Notter et al., 2010).

NMP Recovery

Description	Input	Output	Unit	ecoinvent material/process	Reference/Remarks
Avoided product					
NMP recovered		117.078	g	N-methyl-2-pyrrolidone, at plant/RER U	98% recovery based on LG Chem Ltd. and Compact Power, Inc (U.S. DOE, 2010)
Others					
Water for NMP recovery	9557		g	Water, decarbonised, at plant, RER U	Geisler et al., 2004
Nitrogen for NMP recovery	2		g	Nitrogen, via cryogenic air separation, production mix, at plant, gaseous EU-27 S	Geisler et al., 2004
Steam for NMP recovery	176		g	Steam, for chemical processes, at plant/RER U	Geisler et al., 2004
Energy and processes					
Electricity for NMP recovery	0.006		KWh	Electricity, medium voltage, at grid/US U	Geisler et al., 2004

Table 5. Input-output table for NMP recovery post-anode production

1.3 Electrolyte

Description	Input	Output	Unit	ecoinvent material/process	Reference/Remarks
Functional unit					
Electrolyte		117.52	g		ANL, 2011 (Also volume=0.10 liter)
Materials					
Lithium hexafluorophosphate	17.85		g	Lithium hexafluorophosphate, at plant/CN U	1.20 mol per liter(Nelson et al., 2011)
Ethylene carbonate	99.67		g	Ethylene carbonate, at plant/CN U	Calculated as remaining mass of electrolyte
Transport					
Rail Transport	0.0705		tkm	Transport, freight, rail/RER U	ecoinvent standard distances
Road Transport	0.01175		tkm	Transport, lorry >16t, fleet average/RER U	ecoinvent standard distances

Table 6. Input-output table for the production of electrolyte

LIB electrolyte is composed of lithium hexafluorophosphate (electrolyte salt) and ethylene carbonate (electrolyte solvent). The mass and volume of electrolyte per cell was obtained from the BatPac model, based on the defined cell parameters. Concentration of the electrolyte salt was

obtained from Nelson et al. (2011) and subsequently, the individual masses of the salt and solvent were calculated.

1.4. Separator

Description	Input	Output	Unit	ecoinvent material/process	Reference/Remarks
Functional unit					
Separator		13.9	g		ANL (2011)
Materials					
Polypropylene (PP)	11.12		g	Polypropylene, granulate, at plant/RER U	4:1 ratio of PP and PE based on Nelson et al. (2011)
Polyethylene (PE)	2.78		g	Polyethylene, LDPE, granulate, at plant/RER U	4:1 ratio of PP and PE based on Nelson et al. (2011)
Energy and processes					
Separator production	13.9		g	Injection moulding/RER U	ecoinvent Centre (2010)
Transport					
Rail Transport	2.8E-3		tkm	Transport, freight, rail/RER U	ecoinvent standard distances
Road Transport	1.4E-3		tkm	Transport, lorry >16t, fleet average/RER U	ecoinvent standard distances
Infrastructure					
Facility	1.03E-10		p	Plastics processing factory/RER/I U	ecoinvent standard dataset

Table 7. Input-output table for the production of LIB separator

The composition of LIB separator was obtained from ANL BatPaC model (ANL, 2011). The separator in this model is a 20 micron thick, trilayer PP/PE/PP microporous membrane, which is produced by a "dry" process (Arora and Zhang, 2004). The plastic injection moulding process is used as proxy for separator production (Ellingsen et al., 2013).

1.5. Cell Assembly

Description	Input	Output	Unit	ecoinvent material/process	Reference/Remarks
Functional unit					
LIB single cell		819.85	g		ANL (2011)
Materials					
LIB cathode	402.14		g		ANL (2011)
LIB anode	213.26		g		ANL (2011)
Electrolyte	117.52		g		ANL (2011)
Separator	13.9		g		ANL (2011)
Positive terminal assembly (Aluminum tab)	7.31		g	Aluminium, production mix, at plant/RER U	ANL (2011)
Negative terminal Assembly (Copper tab)	24.14		g	85% Copper, primary, at refinery/RER U; 15% Copper, primary, at refinery/RER U	ANL (2011)
Aluminum in cell container	34.01		g	Aluminium, production mix, at plant/RER U	Estimated from Al layer thickness and density
PET in cell container	5.29		g	Polyethylene terephthalate, granulate, amorphous, at plant/RER U	Estimated from PET layer thickness and density from ANL (2011)
Polypropylene in cell container	2.27		g	Polypropylene granulate (PP), production mix, at plant RER	Estimated from PP layer thickness and density from ANL (2011)
Energy and processes					
Aluminum tab production	7.31		g	Sheet rolling, aluminium/RER U	ecoinvent Centre (2010)
Copper tab production	24.14			Sheet rolling, copper/RER U	ecoinvent Centre (2010)
Calendering cathode, separator and anode	0.0016		KWh	Electricity, medium voltage, at grid/US U	Based on Notter et al. (2010)
Process heat	0.086		MJ	Heat, natural gas, at industrial furnace >100kW/RER U	Heating of cathode, separator and anode. Based on specific heat of materials
Electricity for dry room	0.2		kWh	Electricity, medium voltage, at grid/US U	Dunn et al. (2012)
Natural gas for dry room	1.35		MJ	Natural gas, burned in industrial furnace >100kW/RER U	Dunn et al. (2012)
Aluminum foil production for cell container	34.01		g	Sheet rolling, aluminium/RER U	ecoinvent Centre (2010)
Plastic production for cell container	7.56		g	Injection moulding/RER U	ecoinvent Centre (2010)
Single cell charge, 70%	0.087		kWh	Electricity, medium voltage, at grid/US U	ecoinvent Centre (2010)
Transport					
Rail Transport	1.46E-2		tkm	Transport, freight, rail/RER U	ecoinvent standard distances
Road Transport	7.31E-3		tkm	Transport, lorry >16t, fleet average/RER U	ecoinvent standard distances
Infrastructure					
Facility	3.3E-10		p	Chemical plant, organics/RER/I U	ecoinvent standard dataset

Table 8. Input-output table for LIB cell assembly

Prior to cell assembly, the cathode, separator and anode are heated at 130 degree C to remove any additional moisture (Notter et al., 2010). During cell assembly, cathode, separator and anode are calendared and slit to size-a process requiring electricity (Notter et al., 2010).

The cell housing material is a tri-layer consisting of an outer layer of 0.03 mm polyethylene terephthalate (PET), a middle layer of 0.1 mm aluminum and a 0.20 mm inner layer of polypropylene (PP) (ANL, 2011). The mass of each of these materials in the cell container is estimated from their respective thickness and material densities. The cell assembly takes place in a dry room, energy consumption for which is estimated from Dunn et al. (2012).

S.1.2. LIB Pack

Each LIB pack was modeled to consist of 192 cells, weighing approximately 157 kg based on the contribution of cell weight to the total battery weight as estimated from the BatPac model (ANL, 2011). The remaining weight of the battery pack assembly was composed of the pack and module components, such as module casing, connectors and wiring, battery management system (BMS), battery jacket, and coolant material.

Battery pack assembly component	**Weight (kg)**	**Comments**
Battery pack components	28.63	Includes battery jacket, module compression plates and steel straps and module interconnects (Cu)
Module components	27.49	Includes module casing, module terminals (Cu), cell group interconnects (Cu) and conductor plates (Al)
Battery Management System (BMS)	4.77	Includes the weight of Pack Integration Unit and Module State-of-Charge Regulator Assembly
Battery coolant	4.80	50% Ethylene Glycol and 50% Water (Nelson et al., 2011)
LIB cells	157.41	192 LIB cells
Total Battery weight	223.17	Single EV LIB pack

Table 9. EV LIB pack assembly components

EV LIB pack components

Description	Input	Output	Unit	ecoinvent material/process	Reference/Remarks
Functional unit					
Battery pack components		28.63	kg		ANL (2011)
Materials					
Battery jacket aluminum layers	10.72		kg	Aluminium, production mix, at plant/RER U	Estimated from the thickness and density of Aluminum layer of battery jacket based on ANL (2011) and Nelson et al. (2011)
Battery jacket insulation layer	11.32		kg	Glass fibre, at plant/RER U	Estimated from data from ANL (2011) and Nelson et al. (2011)
Module compression plates and steel straps (steel)	4.20		kg	Steel, low-alloyed, at plant/RER U	ANL (2011)
Module interconnects (Cu)	2.40		kg	85% Copper, primary, at refinery/RER U; 15% Copper, primary, at refinery/RER U	ANL (2011); Material choice based on Nelson et al. (2011)
Energy and processes					
Aluminum jacket production	10.72		kg	Sheet rolling, aluminium/RER U	ecoinvent Centre (2010)
Insulation manufacturing	11.32		kg	Injection moulding/RER U	Proxy for insulation production
Compression plates and straps production	4.20		kg	Steel product manufacturing, average metal working/RER U	ecoinvent Centre (2010)
Copper connector production	2.40		kg	Copper product manufacturing, average metal working/RER U	ecoinvent Centre (2010)
Transport					
Rail Transport	5.726		tkm	Transport, freight, rail/RER U	ecoinvent standard distances
Road Transport	2.863		tkm	Transport, lorry >16t, fleet average/RER U	ecoinvent standard distances
Infrastructure					
Facility for metal components production	7.93E-9		p	Metal working factory/RER/I U	ecoinvent standard dataset
Facility for insulation production	8.38E-9		p	Plastics processing factory/RER/I U	ecoinvent standard dataset

Table 10. Input-output table for the production of EV LIB pack components

The EV battery pack components consist of battery jacket, module compression plates and steel straps and module interconnects. The battery jacket is comprised of two layers of aluminum with insulation material (fiber glass) sandwiched in between (Nelson, 2011). While the jacket mass was obtained from the BatPac model, the mass of aluminum in the jacket was calculated from the dimensions of the pack and thickness and density of the aluminum layer. The insulation mass was simply estimated by subtracting the weight of the Al layer from the overall jacket mass.

EV LIB Module Components

Description	Input	Output	Unit	ecoinvent material/process	Reference/Remarks
Functional unit					
Module components		27.49	kg		ANL (2011)
Materials					
Module casing	9.8208		kg	Aluminium, production mix, at plant/RER U	ANL (2011)
Module terminals	1.8768		kg	85% Copper, primary, at refinery/RER U; 15% Copper, primary, at refinery/RER U	ANL (2011)
Cell group interconnects	0.9984		kg	85% Copper, primary, at refinery/RER U; 15% Copper, primary, at refinery/RER U	ANL (2011)
Conductor plates	14.784		kg	Aluminium, production mix, at plant/RER U	ANL(2012); Material choice based on Nelson et al. (2011)
Energy and processes					
Module casing manufacture	9.8208		kg	Sheet rolling, aluminium/RER U	ecoinvent Centre (2010)
Module terminals manufacture	1.8768		kg	Copper product manufacturing, average metal working/RER U	ecoinvent Centre (2010)
Cell group interconnects manufacture	0.9984		kg	Copper product manufacturing, average metal working/RER U	ecoinvent Centre (2010)
Conductor plates manufacture	14.784		kg	Sheet rolling, aluminium/RER U	ecoinvent Centre (2010)
Transport					
Rail Transport	5.498		tkm	Transport, freight, rail/RER U	ecoinvent standard distances
Road Transport	2.749		tkm	Transport, lorry >16t, fleet average/RER U	ecoinvent standard distances
Infrastructure					
Facility for metal components production	1.26E-8		p	Metal working factory/RER/I U	ecoinvent standard dataset

Table 11. Input-output table for the production of EV LIB pack components

Table 11 represents the overall materials used in the 48 modules within the EV LIB pack comprising 27.49 kg. This includes the aluminum cooling plates (conductor plates) that are provided for thermal management to avoid overheating of LIB cells (Nelson et al., 2011). Due to safety concerns, the coolant liquid (ethylene glycol and water) cannot be in direct contact with the LIB cells, and therefore these plates are provided between the cells, which are directly cooled by this mixture.

EV LIB pack assembly

Description	Input	Output	Unit	ecoinvent material/process	Reference/Remarks
Functional unit					
EV LIB pack		223.17	kg		
Materials					
Battery pack components	28.63		kg		Table 10
Module components	27.49		kg		Table 11
Battery Management System (BMS)	4.77		kg	Modeled in ecoinvent based on input-output data from Ellingsen et al. (2014)	Calculated from BMS composition in Supporting Information of Ellingsen et al. (2014), section 2.4
Ethylene glycol in coolant	2.4		kg	Ethylene glycol, at plant/RNA	50% by weight in coolant (Nelson et al., 2011)
Water in coolant	2.4		kg	Water, deionised, at plant/CH U	50% by weight in coolant (Nelson et al., 2011)
LIB cells	157.41		kg		Table 8
Energy and processes					
Welding	0.089		kWh	Electricity, medium voltage, at grid/US U	Supporting information for Ellingsen et al. (2014) table S2
Testing/activating	16.8		kWh	Electricity, medium voltage, at grid/US U	Assumption of 70% battery charging
Transport					
Rail Transport (coolant)	2.922		tkm	Transport, freight, rail/RER U	ecoinvent standard distance for organic chemicals
Road Transport (coolant)	0.487		tkm	Transport, lorry >16t, fleet average/RER U	ecoinvent standard distance for organic chemicals
Infrastructure					
Facility	1.02E-7		p	Metal working factory/RER/I U	ecoinvent standard dataset

Table 12. Input-output table for EV LIB pack assembly

The various pack components and cells along with their modules are assembled together to constitute a single EV LIB pack. A BMS is provided for monitoring battery state of charge and thermal performance as well as for cell balancing (Nelson et al., 2011). BMS modeling was based on data from a recent LCA study by Ellingsen et al. (2014). A 50/50 ethylene glycol and deionized water mixture (by weight) was assumed for battery coolant material owing to its low

cost and common usage (ANL, 2011; Nelson et al., 2011). This mixture is added for thermal management to ensure cooling between LIB cells by providing a medium to reject heat from the cells. This is achieved by cooling the module conductor plates (aluminum) which are in direct contact with the cells (described in the previous section). Electricity usage for welding during LIB pack assembly process was calculated using data from Ellingsen et al. (2014). Additionally, electricity input was included to account for testing and activating the EV LIB pack (Notter et al., 2010).

S.2. EV battery use

The lifespan of the lithium-ion battery in the EV is assumed to be 8 years which is consistent with many vehicle manufacturers' warranty terms such as Honda and Nissan. The EV use phase of the LIB was modeled as the electricity lost due to battery efficiency over the lifetime of the EV and the additional energy needed to carry the weight of the battery. This approach to model the use phase of EV battery has been used in previous LCA studies (Van den Bossche *et al*., 2006; Zackrisson *et al*., 2010).

Charge-discharge energy efficiency loss: Battery efficiency determines the amount of energy taken out during discharge after the battery was initially charged. According to Rydh and Sandén (2005), the efficiency of lithium-ion batteries can lie anywhere between 85% and 95%. A constant battery efficiency is generally assumed over the entire EV use phase of the battery (Van den Bossche *et al*., 2006; Zackrisson *et al*., 2010). However, the decay in capacity of EV battery is accompanied with increased efficiency loss over its lifetime (Andersson et al., 2002). While capacity fade of LIBs with cycling has been vastly investigated (Arora et al., 1998; Spotniz, 2003), not much has been published about the corresponding phenomenon of energy efficiency fade. In this study, it is assumed that efficiency fade of LIB during EV use phase exhibits direct correlation with capacity fade. Moreover, a linear trend in efficiency fade of LIB has been considered on a daily basis over a period of 8 years (i.e. 2,920 days), assuming that the battery is cycled daily. Similar assumption of linear decrease in EV LIB efficiency with cycling has been assumed in previous studies (Ahmadi et al., 2014a, 2014b). At the beginning of life of EV LIB, a roundtrip efficiency of 95% has been assumed based on advanced vehicle tests for Nissan Leaf battery (Garetson, 2013). At EV end-of-life, when the residual capacity is 80%, the battery efficiency is also considered to be 80% (Ahmadi et al., 2014b). The efficiency has been modeled to decrease linearly with a constant decline of 5.13E-5.

A constant distance travelled by the EV every day (55 km) and a fixed rate of daily decline in battery efficiency is considered over the LIB lifespan in EV. The charge-discharge electricity loss due to battery efficiency (E_{loss}) was calculated from the distance travelled by the BEV over its lifetime (D_{EV}), LIB service life in EV (l_{EV}), number of days of EV use per year (d_{EV}), LIB efficiency at day *t* of its use (η_t), and energy consumption rate of EV (R_{EV}):

$$E_{loss} = \sum_{t}^{(l_{EV} * d_{EV})} ((D_{EV} / (l_{EV} * d_{EV})) * (1 - \eta_t) * R_{EV}) / \eta_t \qquad (1)$$

Values of these parameters are listed in Table 13:

Parameter	Value	Reference/Comments
EV battery weight (W_b)	223 kg	ANL (2011)
Curb weight of EV (W_{EV})	1,456 kg	Includes weight of BEV and LIB. Typical BEV weight without battery (1,233 kg) was estimated from average of Nissan Leaf curb weight (1,508 kg)[1] by subtracting the weight of a 275 kg LEAF battery (Nissan North America, Inc., 2013) from it.
Percent of electricity consumed by the EV due to curb weight of the vehicle (PE_{EV})	30%	Zackrisson (2010)
Energy consumption rate of EV (R_{EV})	0.16 kWh/km	ANL (2011) [Based on BatPac model parameter]
Distance travelled by the BEV over its lifetime (D_{EV})	160,934.4 km (i.e. 100,000 miles)	Functional unit of Case 1 of LCA model– Based on Nissan Leaf battery warranty terms (Nissan North America, Inc., 2014)
LIB service life in EV (l_{EV})	8 years	Based on Nissan Leaf battery warranty terms (Nissan North America, Inc., 2014)
Number of days of EV use per year (d_{EV})	365	Assumed daily use
LIB efficiency in day t (η_t)	95% to 80% for t = 1 to 2,920	Garetson (2013); Ahmadi et al. (2014a,b)
Charging efficiency (η_c)	90%	Rydh and Sanden (2005)

Table 13 Parameters used to calculate EV LIB use phase electricity consumption and losses

Energy to carry EV LIB weight: The additional electricity required to carry the weight of the 223 kg battery (E_b) over the lifetime of the EV was calculated from the ratio of battery weight (W_b) and curb weight of EV (W_{EV}), percent of electricity consumed due to curb weight of the EV (PE_{EV}), distance travelled by the BEV over its lifetime (D_{EV}), charging efficiency (η_c), and energy consumption rate of EV (R_{EV}):

$$E_b = (W_b / W_{EV}) * PE_{EV} * D_{EV} * (R_{EV} / \eta_c) \qquad (2)$$

Values of these parameters are listed in Table 13. The ratio (R_{EV} / η_c) represents the plug-to-wheel consumption rate of the vehicle.

In addition to electricity consumption, the transport of the battery from the battery manufacturer to the car assembly site has been included in the use phase. ecoinvent transport dataset based on Borken-Kleefeld and Weidema (2013) for shipping "other" electronic and electrical equipment has been used as proxy, and included road and transoceanic freight. The LCA inputs for the use phase of a 223 kg EV battery are shown in Table 14

[1] http://www.nissanusa.com. Accessed 15 May 2013

Inputs	Value	Reference/Comments
Electricity used	1,314.41 kWh	Electricity needed to carry battery weight. Based on equation 2
Electricity losses	3,750 kWh	Charge-discharge losses based on battery efficiency. Based on equation 1
Transport, road	71.4 tkm	Borken-Kleefeld and Weidema (2013)
Transport, transoceanic freight ship	248.2 tkm	Borken-Kleefeld and Weidema (2013)

Table 14. Inputs for modeling the use phase of EV LIB

S.3. EV LIB refurbishment

To be refurbished, EV batteries will have to be collected from vehicle dealerships or service centers, inspected and tested to determine their working condition and electrical performance, and reconfigured into battery packs suitable for stationary applications (Cready et al., 2003). The following assumptions have been made for the battery refurbishment stage:

1) EOL EV batteries will be transported to a refurbishment facility. There is immense uncertainty regarding the distance travelled by EOL EV batteries to the refurbishment facility, as currently there are no such facilities under operation. To a large extent it would depend on the total number of such facilities operating in a specific state as illustrated in Cready et al. (2003). In a recent study, Neubauer et al. (2012) have assumed a distance of 30,000 miles (i.e.48,280 km) for transport of 115,920 kWh of used EV LIBs to a refurbishing facility. Considering battery energy density of 108 Wh/kg for the modeled EV battery from the BatPac model, 1 metric tonne of used EV batteries would be transported over a distance of 45 km. Table 15 show the resultant transport burdens for shipping EV LIBs needed to build a single stationary battery system (100% cell conversion, not considering subsequent replacements)

Description	Input	Output	Unit	ecoinvent process	Reference/Remarks
Battery transport to refurbishment facility		5230.55	kg		Transport of 23.44 EV LIB packs to build a single stationary battery pack storing 450 kWh energy
Road Transport	235.35		tkm	Transport, lorry >16t, fleet average/RER U	Neubauer et al. (2012)

Table 15. Transport input for shipping 23.44 EV LIB packs to build a single stationary battery pack storing 450 kWh energy

2) The LIB cells would be tested for their remaining capacity in the refurbishment facility. Preliminary bench scale test results were used to estimate electricity required for testing the LIB cells using a Maccor battery test bed. The cells tests consisted of 4 charge/discharge cycles: Charging according to manufacturer profile while discharging at C/3 to 100% rated capacity (Cready et al., 2003). Table 16 shows the electricity used in testing 4,500 cells that was estimated from bench scale tests (to build a single stationary battery pack with 100% cell conversion and no battery replacement).

Description	Input	Output	Unit	ecoinvent process	Reference/Remarks
LIB cell testing		3689.3	kg		4,500 EV LIB cells comprising a 450 kWh stationary battery pack
Electricity usage	4811.957		kWh	Electricity, medium voltage, at grid/US U	Estimated from bench scale testing of LIB cells

Table 16. Electricity inputs for testing 4,500 cells from EV LIB packs to build a single stationary battery pack storing 450 kWh energy (100% cell conversion, considering no subsequent replacements)

3) After the lithium-ion cells are tested, based on the cell conversion rate some of them would be considered unfit for secondary application and would be shipped to a recycling facility. The feasible cells would be repackaged into modules. It is assumed that the battery pack components such as the battery jacket and BMS would be separated and sent for waste management. However, most of the module components would be reused in building the stationary energy pack. For module assembly, copper connectors and electricity inputs for welding will be required, as shown in Table 17.

Description	Input	Output	Unit	ecoinvent materials/process	Reference/Remarks
Module assembly	4,333.55	4,389.80	kg		Input includes EV LIB cells and module components. Output includes mass of EV LIB cells, EV LIB modules and interconnects in a 450 kWh stationary battery pack
Materials					
Module interconnects	56.25		kg	85% Copper, primary, at refinery/RER U; 15% Copper, primary, at refinery/RER U	Estimated from module interconnects used in EV LIB pack (ANL, 2011)
Energy and processes					
Module interconnects production	56.25		kg	Copper product manufacturing, average metal working/RER U	ecoinvent Centre (2010)
Welding	1.756		kWh	Electricity, medium voltage, at grid/US U	Supporting information for Ellingsen et al. (2014) table S2
Transport					
Rail Transport	11.25		tkm	Transport, freight, rail/RER U	Transport associated with module interconnects production. ecoinvent standard distances
Road Transport	5.625		tkm	Transport, lorry >16t, fleet average/RER U	Transport associated with module interconnects production. ecoinvent standard distances
Infrastructure					
Refurbishment facility	2E-06		p	Metal working factory/RER/I U	ecoinvent standard dataset

Table 17. Module assembly for refurbished LIB based stationary energy storage system

4) Additional components would be added such as battery cabinet and BMS. These are likely to be set up at the site of installation of the stationary energy storage system. A simplifying assumption made here is that the environmental impacts (hence, mass) of the BMS per unit cell for the stationary energy storage system would be half of the mass of BMS per unit cell for the EV battery pack. This assumption is based on the fact that the

BMS for a BEV battery pack will be more robust with advanced circuitry as compared to the BMS for the less demanding stationary battery. The BMS is modeled based on composition data from Ellingsen et al. (2014). For a typical stationary battery comprising of 4,500 EV LIB cells, the BMS would weigh about 56 kg. It has been assumed that during the 20 year lifespan of the stationary battery system, there will be a single replacement of the BMS. The battery cabinet is assumed to be constructed of mild steel sheet (European Telecommunications Standards Institute [ETSI], 2012) with dimensions 114 cm*180 cm* 170 cm and thickness of 0.2 cm. This volume has been estimated from dimensions of a single module obtained from BatPac model. The actual cabinet dimensions would depend upon how the modules are assembled in the stationary battery. The weight of the cabinet was estimated at 221 kg based on density of mild steel (7.85 g/cm^3). The cabinet is assumed to have a 20 year lifespan.

Description	Input	Output	Unit	ecoinvent material/process	Reference/Remarks
Functional unit					
Steel cabinet		1.00	kg		
Materials					
Mild steel	1.00		kg	Steel, low-alloyed, at plant/RER U	Material selection based on ETSI (2012). [Proxy for galvanized steel]
Energy and processes					
Cabinet production	1.00		kg	Sheet rolling, steel/RER U	ecoinvent Centre (2010)
Transport					
Rail Transport	0.2		tkm	Transport, freight, rail/RER U	ecoinvent standard distances
Road Transport	0.1		tkm	Transport, lorry >16t, fleet average/RER U	ecoinvent standard distances
Infrastructure					
Cabinet production facility	4.6E-10		p	Metal working factory/RER/I U	ecoinvent standard dataset

Table 18 Steel cabinet manufacturing for stationary LIB

S.4. Stationary battery use

For the stationary energy storage case (Case 2), the basic assumption is that both Li-ion and PbA batteries have energy storage of 450 kWh (hence, the product, Voltage * Capacity is equal for them). Both batteries deliver 150 kWh everyday-this was calculated for each cycle by adjusting the depth of discharge (DoD) (which is in the range of 33-42%) as the batteries aged. Currently, there is immense uncertainty in sizing an old LIB relative to an existing PbA battery. Owing to performance uncertainties of the former, this sizing approach is considered to be a reasonable extreme sizing scenario for an energy storage system based on second use EV LIBs. A single EV battery (new) modeled in BatPac is assumed to have a peak power of 80 kW which is similar to a Nissan LEAF battery (Garetson, 2013). At end of its service life in EV, the nominal as well as peak power capability of EV LIB is expected to decrease by at least 25% from its initial value due to increased resistance with aging (Burke, 2009). The PbA battery in Rydh (1999) upon which the modeled PbA battery in this study is primarily based has a nominal power rating of 50 kW, which is suitable for the energy storage application selected for this study (Refer to Chapter

3). Hence, the stationary energy storage system based on second use LIBs is more than likely to meet this power requirement.

S.4.1. Refurbished EV LIB based energy storage system (use phase)

The secondary use phase of EV LIBs will include the transport of the refurbished battery to its usage site and also the electricity losses due to the charge-discharge efficiency of the battery system. The total energy used to charge the battery and electricity transmissions losses are outside the system boundary of this study and are considered to be part of the larger grid system.

The transport distance of the refurbished LIB modules for installation in the stationary energy storage system is highly uncertain, and would depend on the locations of these systems as well as the refurbishment facility (Cready et al., 2003). Since, currently these systems don't exist, the default average shipping distance for miscellaneous manufactured goods from Borken-Kleefeld and Weidema (2013) was used for this purpose (0.22 tonne-km/kg). Also, BMS and the cabinet would be shipped and installed at the site. Road based transport has been assumed for modeling the transport of these stationary battery components. The assumptions and proxies used for modeling these transport impacts are provided in Table 19:

Transport description	**Weight transported (kg)**	**Transport input (tonne-km)**	**ecoinvent process**	**Assumption**
Transport of LIB cell, modules and connectors	4389.8	970.15	Transport, lorry >16t, fleet average/RER U	Default data for transportation of miscellaneous manufactured products (Borken-Kleefeld and Weidema, 2013)
BMS	56	26.38	Transport, lorry >16t, fleet average/RER U	Default data for transportation of electronic components and board (Borken-Kleefeld and Weidema, 2013)
Steel cabinet	221	80.30	Transport, lorry >16t, fleet average/RER U	Default data for transport of articles of base metals (Borken-Kleefeld and Weidema, 2013)

Table 19 Input data for modeling transport of LIB modules and ancillary components to the stationary energy storage system site.

The charge-discharge electricity losses from operation of the stationary battery is estimated from the energy storage capacity of the battery at the start of stationary use (C_s), percent residual capacity of the stationary battery at beginning of a given cycle k (PS_k),charge-discharge energy efficiency of the battery at cycle k (η_k), depth-of-discharge of stationary battery during that cycle (DoD_k) and cycle life of retired EV cells in stationary energy storage system (l_c) :

$$E_{loss,s} = \sum_{k}^{l_c} (C_s * PS_k * (1-\eta_k) * DoD_k) / \eta_k \qquad (3)$$

The round trip energy efficiency of the refurbished LIB based energy storage system is assumed to be 80% at beginning of its service life in stationary application, and declines linearly, reaching 65% (w.r.t initial EV LIB) at stationary battery end-of-life. This is based on a similar assumption by Ahmadi et al. (2014a,b). Only the internal energy efficiency of the battery has been

considered to calculate the use phase losses of the battery. Other losses owing to electricity transmission efficiency and efficiencies of charger and inverter have not been included.

The battery capacity has been modeled to reduce linearly by a constant ranging from 0.23 to 0.023 with stationary LIB lifespan in the range of 1 to 10 years (365 to 3,650 cycles) respectively. The depth of discharge of the battery lies in the range of 33% to 42%.

The initial energy storage capacity (C_s) of a stationary battery resulting from a single EV LIB pack is estimated from the initial energy storage capacity (C_{EV}) of the EV battery (i.e. 24 kWh), percent residual capacity of the LIB at EV end-of-life (PC_{EV}) and the cell conversion rate (*F*):

$$C_s = C_{EV} * PC_{EV} * F \quad (4)$$

The cell conversion rate (*F*) is defined as the percentage of EV LIB cells technically feasible for stationary energy storage use. This calculation is made under the assumption that the EV LIB cells at their end of life would retain 80% of their initial capacity (PC_{EV}) while the cell conversion rate (*F*) is scenario dependent as explained in Chapter 3. For the extended life cycle case (Case 1) of EV LIB, this stationary battery represents only a part of the stationary energy storage system corresponding to the functional unit for the first objective of this LCA study (i.e. one 24 kWh EV LIB). Depending on the stationary application and the size of the stationary battery several retired EV battery packs maybe required to build a single stationary battery pack (Cready et al., 2003).

S.4.2. Stationary lead-acid battery use phase

The stationary use phase of the PbA battery includes its transport to the usage site and also the electricity losses due to charge discharge efficiency of the battery system.

Transport: For transport of PbA batteries to the site of usage, the default average shipping distance for miscellaneous manufactured goods was used (0.22 tonne-km/kg) from Borken-Kleefeld and Weidema (2013). Steel cabinet (Refer to section S6) transport was based on the default shipping distance for articles of base metals (Borken-Kleefeld and Weidema, 2013). Road based transport has been assumed (similar to Table 19 for stationary LIB).

Electricity losses: The charge-discharge electricity losses are calculated in the same way as that for a Li-ion battery based system using equation (3). The efficiency of PbA battery generally lies between 70% and 85% (Rydh and Sanden, 2005; Matheys et al., 2009; Matheys and Autenboer, 2005; Albright et al., 2012; Parker, 2001; Celik et al., 2008; Rydh, 1999). Based on this range, three scenarios were considered for PbA battery efficiency at its beginning of life as explained in Chapter 3. The depth-of-discharge of PbA battery system also lies between 33% and 42% and hence, a corresponding lifespan of 5 years (or 1825 cycles) has been assumed for this battery system (Rydh and Sanden, 2005; Bindner et al., 2005).

Unlike LIBs, PbA batteries do no start with their peak capacity, but rather at a lower capacity at the beginning of life. However, with aging, their capacity increases and for more than half of

their cycle life, they are at a capacity higher than 100% of their nominal capacity and thereafter it declines rapidly until it reaches 80% of its nominal capacity at battery end of life (McDowall, 2000; IEEE, 2011; Bindner et al., 2005). The PbA battery capacity has been modeled to reduce linearly by a constant of approximately 0.05 over its 5 year lifespan. Battery aging is accompanied with increase in resistance and corresponding increase in efficiency losses. A direct correlation between increase in battery internal resistance and increase in efficiency losses is considered. PbA battery resistance generally increases by 25%-50% of its initial value at battery EOL (Albercorp, n.d; Davis et al., 2002). For estimating electricity losses due to PbA battery efficiency over its lifespan, an assumption of 25% increase in resistance and hence a 25% increase in efficiency loss at battery end of life is made. PbA batteries usually fail the standard capacity test when internal resistance rises beyond this (Alber, n.d.). Moreover, this assumption represents the worst case for LIBs when compared with lead-acid batteries. The increase in battery resistance with cycling is more of a linear step function, with a slow increase in the initial cycles and rapid increase in later cycles as illustrated for about 100 cycles by Pavlov and Petkova (2002). A constant linear increase in resistance and hence, linear increase in efficiency loss with cycling is a reasonable assumption for this study, considering the lack of data for as high as 1,825 cycles.

S.5. LIB EOL management

The recycling of cells within an EV battery pack would be occurring at two stages-after being rejected for stationary use and at the end of life of the cells in the secondary energy storage system. Currently both pyrometallurgical and hydrometallurgical processes are being used for recycling spent lithium-ion batteries (Dunn et al., 2012; Fisher et al., 2006). It is assumed that 50% of the EOL LIB cells would be recycled by the pyrometallurgical process (Table 20) while the remaining 50% would be recycled by the hydrometallurgical process (Table 21) based on a recent European Commission study where an equal split between the two recycling routes was considered (Mudgal et al., 2011). The material inputs, transportation distance (tonne km) and energy inputs for these two processes have been obtained from the ecoinvent database (Hischier et al., 2007) which is based on European LIB recycling data from Fisher et al. (2006). The material outputs have been obtained from the bill of materials of the EV battery. It is assumed that only metals would be recovered during the recycling process depending upon their respective recycling efficiencies, while the remaining materials in the LIB cells such as mixed plastics, graphite, electrolyte, binders etc. would be sent to the landfill (Table 20 and Table 21).

Description	Input	Output	Unit	ecoinvent material/process	Reference/Remarks
Waste specification name					
End-of-life LIB cells	1		kg		
Avoided materials					
Manganese oxide		0.35	kg	Manganese oxide (Mn2O3), at plant/CN U	Recovered in the form of MnO_2 (Fisher et al., 2006). Mn recovery estimated from cell BOM and 92% manganese yield (mid value from Wang et al., 2014a). Also, Mn content of MnO_2 is 63% (stoichiometric calculation). Mn_2O_3 used as proxy for MnO_2 since it is the actual input in battery production (Dunn et al., 2012).
Primary aluminum		0.037	kg	Aluminium, primary, at plant/RER U	Estimated from cell BOM, 68% virgin inputs and 55% aluminum yield (mid value from Wang et al., 2014a).
Primary copper		0.012	kg	Copper, primary, at refinery/GLO U	Estimated from cell BOM, 85% virgin inputs and 10% copper yield (mid value from Wang et al., 2014a).
Material inputs					
Water	0.001		m3	Water, unspecified natural origin, US	Hischier et al. (2007), based on Fisher et al. (2006)
Sodium hydroxide	0.35		kg	Sodium hydroxide, 50% in H2O, production mix, at plant/RER U	Hischier et al. (2007), based on Fisher et al. (2006)
Energy and processes					
Electricity	0.8		kWh	Electricity, medium voltage, at grid/US U	Hischier et al. (2007), based on Fisher et al. (2006)
Transport					
Road transport	0.5		tkm	Transport, lorry >16t, fleet average/RER U	Hischier et al. (2007)
Infrastructure					
Recycling facility	5E-10		p	Facilities blister-copper conversion, secondary copper/SE/I U	Hischier et al. (2007)
Waste treatment					
Plastic disposal		0.056	kg	Disposal, plastics, mixture, 15.3% water, to sanitary landfill/CH U	Calculated from cell BOM and includes binder and plastic in cell container and separator
Metal slag		0.2	kg	Disposal, nickel smelter slag, 0% water, to residual material landfill/CH U	Calculated from cell BOM (Al, Cu, Li and Mn). Recovered metal was subtracted
Process residues and other non-recycled materials		0.677	kg	Process-specific burdens, sanitary landfill/CH U	Includes carbon and electrolyte from cell BOM and process inputs to obtain a mass balance of inputs and outputs (water input not included)

Table 20 Pyrometallurgical recycling process for LIB cells

Description	Input	Output	Unit	ecoinvent material/process	Reference/Remarks
Waste specification name					
End-of-life LIB cells	1		kg		
Avoided materials					
Lithium carbonate		0.044	kg	Lithium carbonate, at plant/GLO U	Lithium recovery estimated from cell BOM and 55% lithium yield (mid value from Wang et al., 2014a). Also, Li content of Li2CO3 is 19% (stoichiometric calculation).
Primary aluminum		0.037	kg	Aluminium, primary, at plant/RER U	Estimated from cell BOM, 68% virgin inputs and 55% aluminum yield (mid value from Wang et al., 2014a).
Primary copper		0.012	kg	Copper, primary, at refinery/GLO U	Estimated from cell BOM, 85% virgin inputs and 10% copper yield (mid value from Wang et al., 2014a).
Material inputs					
Water	0.001		m3	Water, unspecified natural origin, US	Hischier et al. (2007), based on Fisher et al. (2006)
Reagent	0.025		kg	Chemicals inorganic, at plant/GLO U	Hischier et al. (2007), based on Fisher et al. (2006)
Sulphuric acid	0.23		kg	Sulphuric acid, liquid, at plant/RER U	Hischier et al. (2007), based on Fisher et al. (2006)
Lime	0.116		kg	Lime, hydrated, packed, at plant/CH U	Hischier et al. (2007), based on Fisher et al. (2006)
Energy and processes					
Electricity	0.14		kWh	Electricity, medium voltage, at grid/US U	Hischier et al. (2007), based on Fisher et al. (2006)
Transport					
Road transport	0.5		tkm	Transport, lorry >16t, fleet average/RER U	Hischier et al. (2007)
Infrastructure					
Recycling facility	4E-10		p	Chemical plant, organics/RER/I U	Hischier et al. (2007)
Waste treatment					
Plastic disposal		0.056	kg	Disposal, plastics, mixture, 15.3% water, to sanitary landfill/CH U	Calculated from cell BOM and includes binder and plastic in cell container and separator
Gypsum (as CaSO4, H2O)		0.339	kg	Disposal, gypsum, 19.4% water, to sanitary landfill/CH U	Hischier et al. (2007), based on Fisher et al. (2006)
Process residues and other non-recycled materials		0.864	kg	Process-specific burdens, sanitary landfill/CH U	Calculated from mass balance of inputs and outputs (water input not included)

Table 21 Hydrometallurgical recycling process for LIB cells

Apart from LIB cell recycling, EOL management of LIB pack and module components was also modeled. In addition, components added to the stationary battery during refurbishment (e.g. BMS, copper connectors) were assumed to be recycled as well at their EOL. The recycling processes for metals in these components were modeled using data for secondary metal production from the ecoinvent database and recycling credit was provided for avoiding the production of primary metal wherever used. Transport of EOL components to a recycling facility

was based on data from Hischier et al. (2007) for waste LIB transport (i.e. 0.5 tonne km of road transport per kg of waste).

Description	Input	Output	Unit	ecoinvent material/process	Reference/Remarks
Waste specification name					
EV LIB pack components EOL management	33.4		kg		Includes battery jacket, module compression plates and steel straps, module interconnects (Cu) and coolant for a single EV LIB pack
Avoided materials					
Primary steel		2.166	kg	Steel, converter, low-alloyed, at plant/RER U	Credit is provided for avoiding 63% primary steel in the initial inputs
Primary aluminum		7.084	kg	Aluminium, primary, at plant/RER U	Credit is provided for avoiding 68% virgin aluminum inputs
Primary copper		1.556	kg	Copper, primary, at refinery/GLO U	Credit is provided for avoiding 85% virgin aluminum inputs
Processes					
Steel recycling	3.8		kg	Steel, electric, un- and low-alloyed, at plant/RER U	Proxy for steel recycling.Material recovery estimated from EV LIB pack BOM and 90% recycling efficiency of steel (based on ecoinvent Centre, 2010)
Aluminum recycling	10.41		kg	Aluminium, secondary, from old scrap, at plant/RER U	Proxy for aluminum recycling. Material recovery estimated from EV LIB pack BOM and 97% yield of secondary aluminum in ecoinvnet database (ecoinvent Centre, 2010)
Copper recycling	1.83		kg	Copper, secondary, at refinery/RER U	Proxy for copper recycling. Material recovery estimated from EV LIB pack BOM and 76% yield for secondary copper in ecoivent database (ecoinvent Centre, 2010)
Transport					
Road transport	16.7		tkm	Transport, lorry >16t, fleet average/RER U	Same as transport data for waste LIBs from Hischier et al. (2007)
Waste treatment					
Disposal of coolant		4.8	kg	Disposal, antifreezer liquid, 51.8% water, to hazardous waste incineration/CH U	LIB pack BOM from ANL (2011)
Disposal of fiber glass insulation in battery jacket		11.32	kg	Disposal, inert waste, 5% water, to inert material landfill/CH U	LIB pack BOM from ANL (2011)

Table 22. EOL management of EV LIB pack components

Table 22 represents the EOL management of EV LIB pack components. In a similar way, EOL management of module components was modeled. The EOL management of BMS was modeled as shown in Table 23.

Description	Input	Output	Unit	ecoinvent material/process	Reference/Remarks
Waste specification name					
EV LIB BMS recycling	4.77		kg		ANL (2011)
Avoided materials					
Primary steel		1.012	kg	Steel, converter, low-alloyed, at plant/RER U	Credit is provided for avoiding 63% primary steel in the initial inputs
Primary aluminum		0.049	kg	Aluminium, primary, at plant/RER U	Credit is provided for avoiding 68% virgin aluminum inputs
Primary copper		0.158	kg	Copper, primary, at refinery/GLO U	Credit is provided for avoiding 85% virgin copper inputs
Processes					
Steel recycling	1.775		kg	Steel, electric, un- and low-alloyed, at plant/RER U	Proxy for steel recycling. Material recovery estimated from BMS BOM (based on Ellingsen et al., 2014) and 90% recycling efficiency of steel (based on ecoinvent Centre, 2010)
Aluminum recycling	0.072		kg	Aluminium, secondary, from old scrap, at plant/RER U	Proxy for aluminum recycling. Material recovery estimated from BMS BOM (based on Ellingsen et al., 2014) and 97% yield of secondary aluminum in ecoinvnet database (ecoinvent Centre, 2010)
Copper recycling	0.186		kg	Copper, secondary, at refinery/RER U	Proxy for copper recycling. Material recovery estimated from BMS BOM (based on Ellingsen et al., 2014) and 76% yield for secondary copper in ecoivent database (ecoinvent Centre, 2010)
Transport					
Road transport	2.385		tkm	Transport, lorry >16t, fleet average/RER U	Same as transport data for waste LIBs from Hischier et al. (2007)
Waste treatment					
Disposal of printed wiring board		2.088	kg	Disposal, treatment of printed wiring boards/GLO U	Includes printed wiring board, IC circuit, passive electronic components and trace amounts of metals like brass and tin. Based on BMS composition data from Ellingsen et al. (2014).
Disposal of plastic parts in BMS		0.4	kg	Disposal, plastics, mixture, 15.3% water, to sanitary landfill/CH U	Based on BMS composition data from Ellingsen et al. (2014).

Table 23. EOL management of BMS from a single EV battery pack.

Similar process modeling was used for EOL BMS for the stationary battery pack but was adjusted for the reduced BMS size. Recycling of copper connectors used in stationary LIB pack was modeled using secondary copper production data from ecoinvent database (ecoinvent Centre, 2010) similar to the processes described in tables above (76% recycling yield, and credit provided for avoiding 85% primary copper). Similarly, recycling process of battery cabinet employed recycling efficiency of 90% and credits were provided for avoiding 63% primary steel inputs (ecoinvent Centre, 2010). Secondary steel production process in ecoinvent database was

used as a proxy for the cabinet recycling process [Steel, electric, un- and low-alloyed, at plant/RER U].

S.6. Lead-acid battery life cycle (production and recycling)

The life cycle of the lead-acid battery system includes the production of the lead-acid battery, its stationary use and its recycling.

The system boundary for Case 1 of this LCA study has been extended to include the life cycle of a PbA battery system used for stationary energy storage which is basically being avoided due to use of retired EV batteries for that purpose (Figure 2.1 in the Chapter 3). A PbA battery storing and delivering energy equivalent to a refurbished EV LIB based stationary system has been assumed. Again, in actuality this system will represent only part of a PbA battery, whose size is determined by the LIB which replaces it (based on cell conversion rate, F of EV LIBs).

For the stationary energy storage case (Case 2), a 450 kWh PbA battery has been considered which provides daily functionality equivalent to a refurbished LIB based system.

S.6.1 Lead-acid Battery production

The material composition of the lead-acid battery was estimated from Rydh (1999), Rantik (1999) and Sullivan and Gaines (2012). Based on an average energy density of 34.5 Wh/kg (Rydh, 1999; Rydh and Sanden, 2005), the mass of the PbA battery system storing 450 kWh of energy was estimated at 13,043.48 kg.

Material	Battery component	Weight percent	Kg/battery	Reference
Lead	Active material, grids and poles	61.20%	7983	Rydh, 1999; Rantik,1999
Water	Electrolyte (dilution to 1.295s.g.)	13.30%	1735	Rydh, 1999
Sulfuric acid	Electrolyte	9.60%	1252	Rydh, 1999
Polypropylene	Cases and cover	10.00%	1304	Sullivan and Gaines, 2012
Antimony (Sb)	Grid alloys	1.00%	130	Sullivan and Gaines, 2012
Glass	Glass mat separator	2.00%	261	Sullivan and Gaines, 2012
Copper	Connector	0.30%	39	Rydh, 1999
Oxygen	In PbO2 (lead oxide)	2.26%	295	Rantik, 1999
Expander	In PbO2(lead oxide)	0.34%	44	Estimated from remaining mass percent

Table 24. Composition of lead-acid battery

A 47/53 mix of recycled and virgin lead is assumed for the PbA battery (Hittman Associates, 1980; Sullivan and Gaines, 2012). In addition to these components, a battery cabinet composed of mild steel is provided for the lead-acid battery system. The amount of steel used in the cabinet was estimated to be 380.44 kg based on lead-acid battery cabinet mass for a solar power system (European Telecommunications Standards Institute [ETSI], 2012), and scaling it on the basis of battery mass for the system defined here. Similar to the stationary LIB system, the cabinet is assumed to have a 20 year lifespan. The production of the battery cabinet has been modeled similar to the cabinet for the PbA system (Table 18). For plastic cases, 70% recycled plastic is assumed, based on the average recycled content for PbA batteries (Battery Council International, 2012). Unlike LIBs, it is not essential to have a BMS for PbA battery systems (Matheys and Autenboer, 2005) and hence it has not been included. The energy use for battery manufacturing

was obtained from Hittman Associates (1980) and included electricity and heat used in the lead oxide paste production, grid manufacturing, plate manufacturing and battery assembly and formation. Table 25 represents the input-output for producing a single PbA battery with a 5 year service life in stationary application [Does not include cabinet manufacturing].

Description	Input	Output	Unit	ecoinvent material/process	Reference/Remarks
Functional unit					
Lead-acid battery system		13043.48	kg		Weight of a PbA battery system
Materials					
Lead	7982.61		kg	53% Lead, primary, at plant/GLO U; 47% Lead, secondary, at plant/RER U	47/53 mix of recycled and virgin lead based on Hittman Associates (1980). Percentage of lead by weight based on Rydh (1999) and Rantik (1999)
Water	1734.78		kg	Water, deionised, at plant/CH U	% by weight based on Rydh (1999)
Sulphuric Acid	1252.17		kg	Sulphuric acid, liquid,at plant/RER U	% by weight based on Rydh (1999)
Polypropylene (Cases and cover)	1304.35		kg	30% Polypropylene, granulate, at plant/RER U; 70% Recycled postconsumer HDPE pellet/RNA	70 percent recycled plastic in the battery based on average percentage from Battery Council International (2012). Postconsumer HDPE used as proxy for recycled polypropylene.
Antimony	130.43		kg	Antimony, at refinery/CN U	Percentage by weight based on Sullivan and Gaines (2012)
Glass	260.87		kg	Glass fibre, at plant/RER U	Percentage by weight based on Sullivan and Gaines (2012)
Copper connectors	39.13		kg	85% Copper, primary, at refinery/RER U; 15% Copper, primary, at refinery/RER U	Percentage by weight based on Rydh (1999)
Oxygen	294.78		kg	Oxygen, liquid, at plant/RER U	Percentage by weight based on Rantik (1999)
Barium sulphate	41.02		kg	Chemicals inorganic,at plant/GLO U	92.5% of barium sulphate in expander based on Boden (1998)
Carbon black	3.33		kg	Carbon black, at plant/GLO U	Average percentage (7.5%) of carbon in expander based on Boden (1998)
Energy and processes					
Plastic cases and cover production	1304.35		kg	Injection moulding/RER U	ecoinvent Centre (2010)
Copper connectors production	39.13		kg	Wire drawing, copper/RER U	ecoinvent Centre (2010)
Electricity	4043.48		kWh	Electricity, medium voltage, at grid/US U	Combined consumption for paste manufacture, grid manufacture, plate manufacture and assembly and formation. Hittman Associates (1980)
Process heat, Natural gas	35608.7		MJ	Natural gas, burned in industrial furnace >100kW/RER U	Combined consumption for paste manufacture, grid manufacture and plate manufacture. Hittman Associates (1980)
Process heat, Heavy fuel oil	3260.87		MJ	Heat, heavy fuel oil, at industrial furnace 1MW/RER U	Combined consumption for grid manufacture and plate manufacture. Hittman Associates (1980)
Transport					
Rail Transport	3202.17		tkm	Transport, freight, rail/RER U	ecoinvent standard distances
Road Transport	1289.6		tkm	Transport, lorry >16t, fleet average/RER U	ecoinvent standard distances
Infrastructure					
Facility for battery assembly	5.22E-6		p	Chemical plant, organics/RER/I U	ecoinvent standard dataset
Facility for active material, grids and poles manufacturing	3.66E-6		p	Metal working factory/RER/I U	ecoinvent standard dataset

Table 25 Input-output table for lead-acid battery production

S.6.2. Lead-acid battery recycling

For the recycling of PbA battery, the energy and material inputs were estimated from lead-acid battery recycling data from Fisher et al. (2006) as well as the ecoinvent data for secondary lead production (ecoinvent Centre, 2010 based on Fisher et al. 2006) which is assumed to have 100% material recovery efficiency. Recycling credits are provided for avoiding the production of 53% primary lead (Table 26).

Description	Input	Output	Unit	ecoinvent material/process	Reference/Remarks
Waste specification name					
Lead-acid battery recycling	1		kg		
Avoided materials					
Primary lead		0.324	kg	Lead, primary, at plant/GLO U	Credit is provided for avoiding 53% primary lead in the battery
Processes					
Lead-acid battery recycling process	0.612		kg	Lead, secondary, at plant/RER U	Proxy for lead-acid battery recycling process. 100% recycling efficiency (Fisher et al., 2006) and 61.2% lead content in the battery is assumed (Rydh, 1999)
Transport					
Road transport	0.5		tkm	Transport, lorry >16t, fleet average/RER U	Same as transport data for waste LIBs from Hischier et al. (2007)

Table 26 Lead-acid battery recycling

Apart from PbA battery recycling, the cabinet was also recycled and appropriate credits provided for avoiding primary steel production. It was modeled similar to recycling of stationary LIB cabinet- recycling efficiency of 90% and credits provided for avoiding 63% primary steel inputs (ecoinvent Centre, 2010). Secondary steel production process in ecoinvent database was used as a proxy for the cabinet recycling process.

S.7. Additional results-Global Warming Potential (GWP)

(A) Extended life cycle (Case 1)

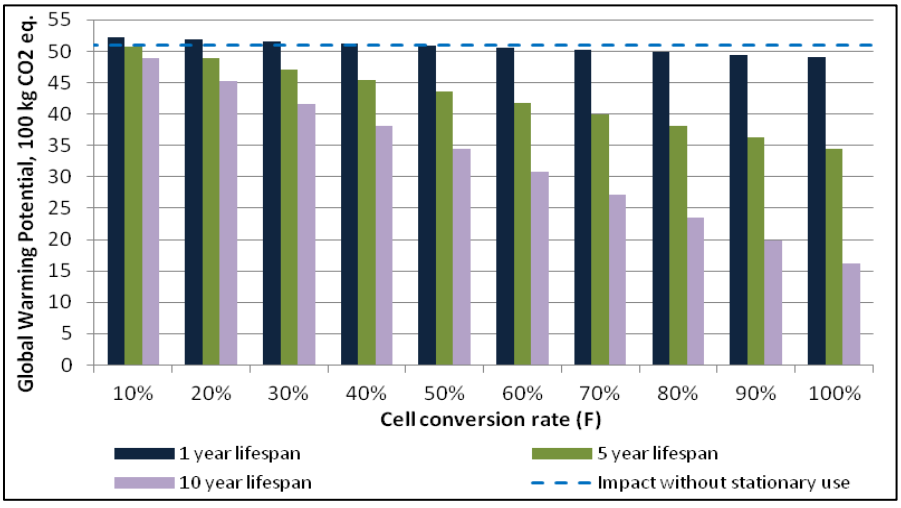


Figure S1: Sensitivity Analysis: Global Warming Potential of EV Lithium-ion battery with extended life under different scenarios of cell conversion rate and refurbished EV LIB lifespan

(C) Stationary energy storage case (Case 2)

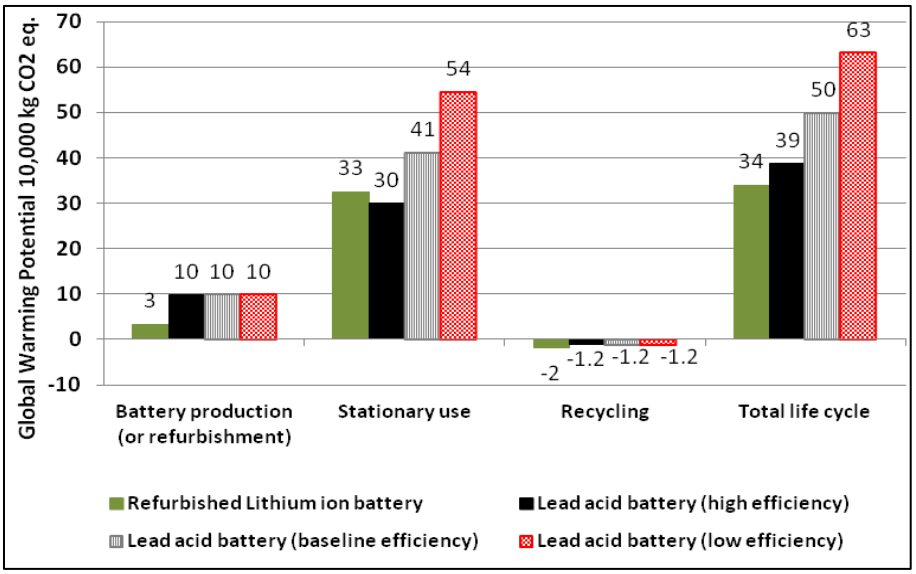


Figure S2: Global Warming Potential of stationary energy storage system (Base case scenario)

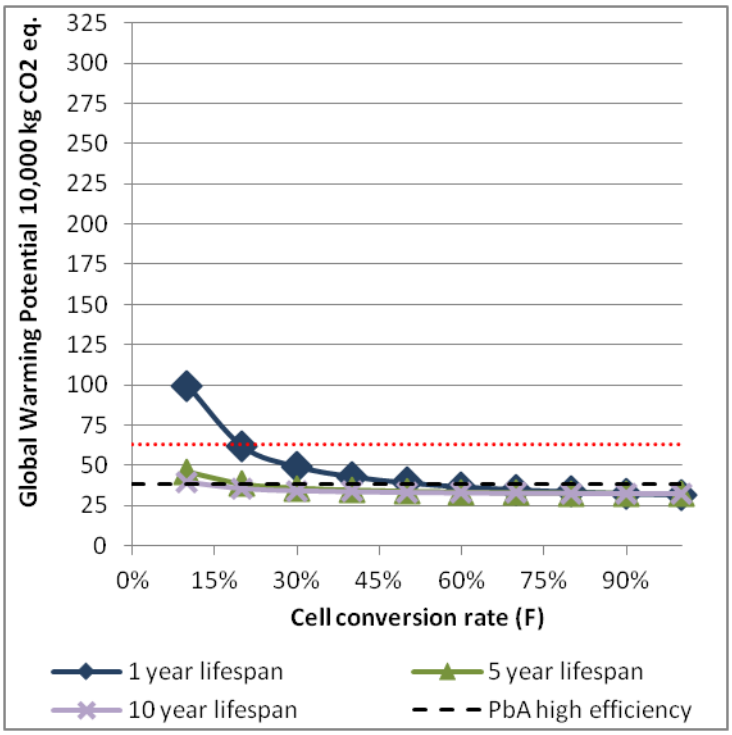


Figure S3: Impact of cell conversion rate and refurbished battery lifespan on environmental feasibility of EV LIB reuse in terms of Global warming potential (cut-off allocation scenario).

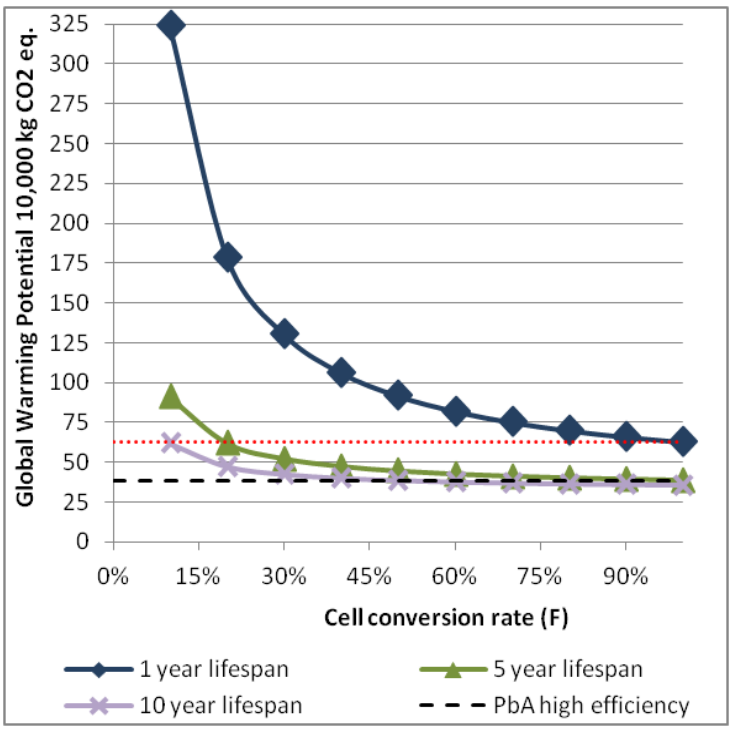


Figure S4: Impact of cell conversion rate and refurbished battery lifespan on environmental feasibility of EV LIB reuse in terms of Global warming potential (50/50 allocation method)

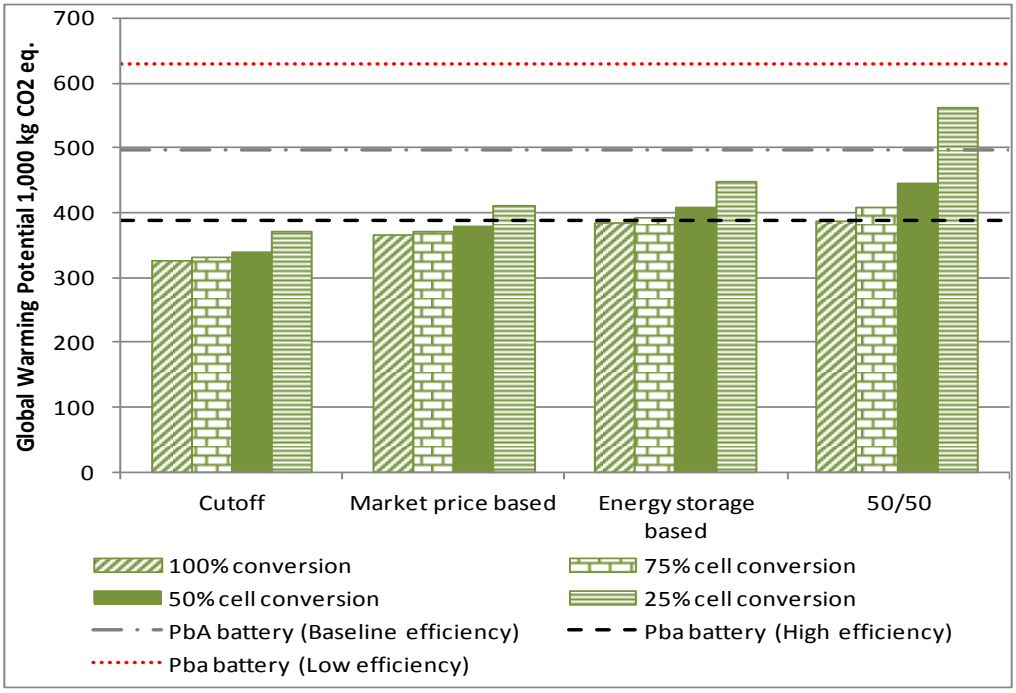


Figure S5: GWP of refurbished EV LIB based energy storage system under different allocation approaches (Considering 5 year battery lifespan in stationary application in all cases)

APPENDIX C

S1. Electric vehicle lithium-ion battery waste flows

Based on a conservative baseline of electric vehicle (EV) adoption, Material Flow Analysis (MFA) results for year 2030 indicated a waste stream of 294,440 lithium-ion battery (LIB) packs (Richa et al., 2014). This forecasted stream consisted of 25% BEV [Battery electric vehicle], 36% long range PHEV [Plug-in hybrid electric vehicle] and 39% short range PHEV LIB packs.

EV-type	BEV	PHEV-50	PHEV-12
Battery units with remaining useful life in EV application	22,838	41,756	45,052
Battery units reaching EOL in EV use	50,841	64,802	69,153

Table S1 (a). EOL LIBs from battery and plug-in electric vehicles-Year 2030. The no. of packs are obtained from the baseline EOL EV battery outflows from Richa et al. (2014).

These forecasted waste flows were normalized to 1,000 LIB packs to indicate a functional unit representing future annual EV LIB waste stream for a given year "n". The mass of the waste stream was calculated from battery packs modeled Argonne National Laboratory BatPac tool (Nelson et al., 2011) resulting in an estimated waste stream of 160 MT for the chosen functional unit.

EV-type	BEV	PHEV-50	PHEV-12	Total
Battery units with remaining useful life in EV application	78	142	153	372
Metric tons	26	23	7.6	56
Battery units reaching EOL in EV use	173	220	235	628
Metric tons	57	35	11.6	104

Table S1 (b). EOL LIBs from battery and plug-in electric vehicles normalized to 1,000 pack units. The mass of the waste stream was calculated from battery packs modeled in Argonne National Lab BatPac model (Nelson et al., 2011).

S2. Battery bill of materials

Individual LIB packs and corresponding cells for BEV and PHEV batteries for the case study were modeled in Argonne National Laboratory BatPac model as indicated in Table S2.

Amount per cell (g)	BEV pack	Low range PHEV pack	High range PHEV pack
Aluminum	79.84	87.01	74.23
Copper	109.18	125.88	101.79
Lithium	12.05	10.83	11.12
Manganese	190.76	171.44	176.07
Graphite	116.49	104.74	107.61
Carbon	21.16	19.02	19.53
Binder	23.76	21.36	21.94
Plastic	20.95	23.63	19.41
Electrolyte	114.58	109.09	105.82
Other	111.04	99.80	102.49
Total cell	799.81	772.79	740.01

Table S2 (a) Bill of materials of individual Lithium Manganese Oxide cells in BEV and PHEV battery packs (Based on Nelson et al., 2011).

	Amount per battery pack (kg)		
Material	**BEV (39 kWh)**	**Low range PHEV (4.4 kWh)**	**High range PHEV (18 kWh)**
Aluminum	65.69	9.61	33.26
Copper	37.28	7.96	18.32
Lithium	3.86	0.43	1.78
Manganese	61.04	6.86	28.17
Steel	7.71	4.37	4.48
Graphite	37.28	4.19	17.22
Carbon	6.77	0.76	3.12
Binder	7.60	0.85	3.51
Plastic	7.14	1.29	3.49
PCB*	2.31	1.82	2.03
Fiber glass	14.98	1.76	8.04
Coolant (Water+ethylene glycol)	6.64	1.19	3.34
Electrolyte	36.67	4.36	16.93
Other	35.53	3.99	16.40
Total	330.50	49.45	160.10

Table S2 (b). Bill of materials of BEV and PHEV battery packs comprising of Lithium Manganese Oxide cells (Based on Nelson et al., 2011)

- The printed circuit board (PCB) is considered to be composed of 35% metal with, 18.67% copper, 0.036% gold, 0.01% palladium and 4.13% aluminum, which are the priority four metals from an environmental and economic standpoint for PCB recycling (Wang and Gaustad, 2012)
- The percentage of hexafluorophosphate (LiPF6) salt in the electrolyte is 15% by mass (Nelson et al., 2011). The remaining electrolyte is composed of ethylene carbonate solvent.

S3. Additional material input during reuse in EV

EV LIB type	BEV	PHEV-12	PHEV-50
No. of packs reused in EVs	78	153	142
No. of cells/pack replaced (10%)	32	4	16

Table S3 (a). No. of cells replaced per pack for each EV LIB type

	78 BEV packs (kg)	153 PHEV-12 packs (kg)	142 PHEV-50 packs (kg)	All packs (kg)
Aluminum	198.16	53.25	168.44	419.84
Copper	270.99	77.05	230.96	578.99
Lithium	29.91	6.63	25.23	61.77
Manganese	473.48	104.93	399.51	977.92
Graphite	289.14	64.10	244.17	597.41
Carbon	52.52	11.64	44.31	108.47
Binder	58.98	13.07	49.78	121.83
Plastic	51.99	14.47	44.04	110.50
Electrolyte	284.41	66.76	240.10	591.27
Other	275.62	61.08	232.56	569.26
Total	1,985.20	472.97	1,679.10	4,137.27

Table S3 (b). Additional material input for LIB reuse in EVs for the case study

S4. EV battery charge-discharge efficiency losses

The charge-discharge electricity loss due to EV battery efficiency (E_{loss}) was calculated from the total distance travelled by the two EVs using the LIB (D_{EV}), design lifespan of LIB in EV use (l_d), number of cycles of EV use per year (d_{EV}), LIB efficiency at cycle *t* of its use (η_t), energy consumption rate of EV in kWh/mile (R_{EV}), and percent of miles powered by electricity (P_e):

$$E_{loss} = \sum_{t}^{(l_d * d_{EV})} ((D_{EV} / (l_d * d_{EV})) * (1 - \eta_t) * R_{EV}) * P_e$$

The above equation is adapted from Zackrisson et al. (2010). A direct correlation between capacity decay and battery charge-discharge efficiency has been assumed and after reuse in EVs, the capacity as well as efficiency of LIBs are reduced to 80% (Ahmadi et al., 2014a,b; Richa et al., 2015). It is assumed that the efficiency decay is linear.

Based on warranty terms for Nissan LEAF battery (Nissan North America, Inc., 2014), it was assumed that typically EVs would provide 100,000 miles operation during the design lifespan of LIB (D_{EV}). For a BEV, 100% of the travel miles are powered by electricity. For the 12 mile and 50 mile electric range PHEVs, this percentage was estimated to be 23% and 61%, respectively

by curve-fitting to prior data corresponding to fraction of total vehicle distance and the PHEV range provided by Samaras and Meisterling (2008).

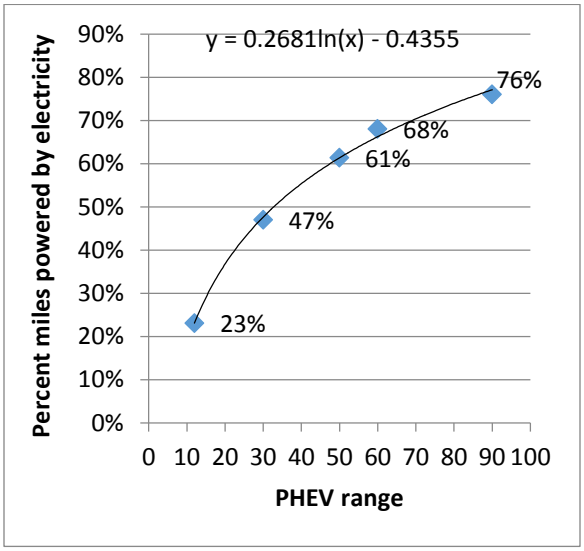


Fig. S4 Fraction of total vehicle distance powered by electricity and the PHEV range

Parameter	Value	Reference/Comments
Energy consumption rate of EV (R_{EV})	0.16 kWh/km	Nelson et al. (2011) [Based on BatPac model parameter]
Distance travelled by the BEV over its lifetime (D_{EV})	160,934.4 km (i.e. 100,000 miles)	Nissan LEAF battery warranty terms (Nissan North America, Inc., 2014)
Design lifespan of EVLIB (l_d)	9 years	Richa et al. (2014)
Number of days of EV use per year (d_{EV})	365	Assumed daily use
LIB efficiency in day *t* (η_t)	95% to 80% for *t* = 1 to 3,285	Garetson (2013); Ahmadi et al. (2014a,b)
Percent of miles powered by electricity (P_e)		12 mile PHEV: 23%; 50 mile PHEV: 61%

Table S4. Parameters used to calculate charge-discharge electricity loss due to battery efficiency (E_{loss}) during EV use

S5. Battery testing for refurbishment for reuse in EVs

Conservative EV LIB testing scenario: One charge at 70% of total battery capacity
Electricity usage (kWh) = Battery capacity*0.70/battery efficiency

Aggressive battery testing scenario: Testing individual cells
Electricity used for testing 1 kWh cell = 10.69 kWh (Richa et al., 2015)
Electricity usage (kWh) =Battery capacity*10.69

The aggressive battery testing scenario is based on bench scale cell testing and includes energy efficiency losses and 4 charge-discharge cycle on a Maccor Test Bed, testing equipment and computer operation over the duration of the test (Richa et al., 2015).

S6. Future new, used and refurbished battery prices

	High ($/kWh)	Low ($/kWh)	Data source
Future New EV battery cost	440	125	Neubauer et al. (2012)
Used battery selling price	100	20	Neubauer et al. (2012)
Refurbished battery buying price	132	38	Neubauer et al. (2012)
Lead acid battery cost	120	65	Albright et al., 2012

Table S6. Future new, used and refurbished battery prices

S7. Environmental Impact of LIB Recycling

LIB cell recycling was modeled from LCI data obtained from the ecoinvent database (Hischier et al., 2007) and Fisher et al. (2006). Through the pyrometallurgical route, manganese is recovered as manganese oxide, while the hyrometallurgical recycling pathway yields lithium carbonate (Fisher et al., 2006). The recycling processes for metals in LIB pack components were modeled using data for secondary metal production from the ecoinvent database.

Data for estimating the environmental impact of LIB recycling is provided in the following table:

	MJ/kg	CTUe/kg	Data source
LIB Cell recycling-hydrometallurgy	5.7	0.8	LCI from Richa et al. (2015), based on Hischier et al., 2007; ecoinvent Centre, 2010; Fisher et al., 2006
LIB Cell recycling-pyrometallurgy	19.6	1.3	LCI from Richa et al.; (2015), based on Hischier et al., 2007; ecoinvent Centre, 2010; Fisher et al., 2007
Steel-secondary	8.9	27.8	ecoinvent Centre, 2010
Aluminum-secondary	23.8	4.47	ecoinvent Centre, 2010
Copper-secondary	28.1	20.9	ecoinvent Centre, 2010
Gold-secondary	7450	223	ecoinvent Centre, 2010
Palladium-secondary	3900	117	ecoinvent Centre, 2010

Table S7 (a). Data for estimating the environmental impact of LIB recycling

Data for estimating the avoided impact from LIB recycling is provided in the following table:

	MJ/kg	CTUe/kg	Data source
Lithium carbonate	42.5	7.5	ecoinvent Centre, 2010
Manganese oxide	37.9	2.3	ecoinvent Centre, 2010
Steel-primary	30.9	8.02	ecoinvent Centre, 2010
Aluminum-primary	194	39.2	ecoinvent Centre, 2010
Iron-primary	25	1.01	ecoinvent Centre, 2010
Copper-primary	60.5	607	ecoinvent Centre, 2010
Nickel-primary	121	17.3	ecoinvent Centre, 2010
Cobalt-primary	128	9.2	ecoinvent Centre, 2010
Gold-primary	313000	981000	ecoinvent Centre, 2010
Palladium-primary	177000	24800	ecoinvent Centre, 2010

Table S7 (a). Data for estimating the avoided impact from LIB recycling

S8. LIB Recycling Cost

Recycling of EOL EV LIBs generated in a given year along with the additional material input for the reuse and cascaded use stages is likely to occur in three cycles C1, C2 and C3 separated by time lags (Figure 4.2 in Chapter 4). Cycle 1 (C1) recycling (year n) would include EV LIBs not tested feasible for reuse or cascaded use in that year, 10% reject cells from EV LIB packs during refurbishment for reuse application in EVs, and rejected pack components during refurbishment and assembly for C1 cascaded use stationary LIB systems. Cycle 2 (C2) recycling would follow 4.5-5 years later and would include EV LIB packs not tested feasible for cascaded use after reuse in EVs, rejected pack components during refurbishment and assembly of C2 cascaded use stationary LIB systems, and C1 cascaded use stationary LIB systems reaching EOL. Cycle 3 (C3) recycling would follow after another 5 years (i.e. year n+9.5) and would include C2 cascaded use stationary LIB systems reaching EOL.

The total cost ($TC_{rec,m}$)of operating LIB recycling facility was calculated using fixed cost of 1,000,000 $/year for a maximum recycling capacity of 34,000 metric tons of LIB waste per year (Wang et al., 2014b). A conservative variable cost of 1,100 $/metric ton of LIB waste was employed (Wang et al., 2014b).

$TC_{rec,m} = 1{,}000{,}000 + (1{,}100*(34{,}000))$

The cost of recycling LIBs for each recycling cycle ($TC_{rec,c}$) was calculated from the weight of LIB waste considered for recycling in each cycle (W_c) for the analyzed waste stream, the maximum recycling capacity (W_m) and the cost of operating the recycling facility ($TC_{rec,m}$):
$TC_{rec,c} = (W_c / W_m) * (TC_{rec,m})$

Total cost of recycling operations (TC_{rec}) for the analyzed waste stream was estimated as the sum of $TC_{rec,c}$ for the three recycling cycles.

S9. LIB metal recycling efficiency and commodity value

Recycled metal	Recycling efficiency [RE] (%)	RE Source	Material price $/kg	Price Source
Aluminum	60	Graedel et al. (2011)	2.08	USGS, 2015
Copper	53	Graedel et al. (2011)	7.33	USGS, 2015
Lithium	55	Mantuano et al. (2006)	6.80 (Lithium Carbonate)	USGS, 2015
Manganese	53	Sibley (2011)	2.22	www.infomine.com
Iron and Steel	52	Graedel et al. (2011)	0.67	USGS, 2015
Nickel	68	Graedel et al. (2011)	15.02	USGS, 2015
Cobalt	57	Graedel et al. (2011)	28.42	USGS, 2015
Gold	96	Graedel et al. (2011)	46,000	USGS, 2015
Palladium	79	Graedel et al. (2011)	23,500	USGS, 2015

Table S9 (a). Metal recycling efficiencies and metal prices

Varying the composition of the analyzed waste stream based on specific chemistry or an equal mix of LMO, NCM and LFP chemistry, resulted in the following estimates for the recycling stream (Considering conservative variable cost of recycling at1,100 $/metric ton):

	LMO	NCM	LFP	Equal mix
Amount of waste sent for recycling (mT)	167	147	189	168
Total cost of recycling (USD)	188,424	166,333	214,130	118,495
Total material value (USD)	237,381	418,323	237,406	226,569
Net economic benefit (USD)	(48,957)	(251,990)	(23,276)	(108,074)
CED (MJ)	(3,521,975.71)	(4,077,610.31)	(4,113,719.73)	(3,904,435.25)

Table S9 (b). Sensitivity analysis of recycling pathway under different LIB chemistry scenario

The material value from the recycling stream was calculated for LFP and NCM chemistry as follows:

1. LFP

Cells recycling

Recycled metal	Amount in waste stream (kg)	% recovery	Actual Recovery (kg)	Material price $/kg	Total recovered value ($)
Aluminum	16,112.46	0.6	9,667.48	2.08	20,076.99
Copper	22,341.34	0.53	11,840.91	7.33	86,746.10
Lithium	1,705.53	0.55	4,937.06	6.80	33,571.99
Iron	13,726.31	0.52	7,137.68	0.67	4,782.25
				Total	145,177.34

Table S9 (c) Material value from LFP cells

Pack material recycling

Recycled metal	Amount in waste stream (kg)	% recovery	Actual Recovery (kg)	Material price $/kg	Total recovered value ($)
Steel	7,436.52	0.52	3,866.99	0.67	2,596.04
Aluminum	24,984.74	0.6	14,990.84	2.08	31,132.33
Copper	4,234.67	0.53	2,244.37	7.33	16,442.21
Gold	0.87	0.96	0.83	45645.16	38,079.11
Palladium	0.24	0.7	0.17	23548.39	3,979.03
				Total	92,228.71

Table S9 (d) Material value from LFP battery pack materials

2. NMC

Cells Recycling

Recycled metal	Amount in waste stream (kg)	% recovery	Actual Recovery (kg)	Material price $/kg	Total recovered value ($)
Aluminum	11,101.63	0.6	6,660.98	2.08	13,833.23
Copper	15,445.75	0.53	8,186.25	7.33	59,972.17
Lithium	2,743.96	0.55	7,943.03	6.80	54,012.61
Manganese	6,550.94	0.53	5,511.11	2.22	12,234.66
Cobalt	7,026.96	0.68	4,778.33	28.42	135,788.70
Nickel	6,998.34	0.57	3,989.05	15.02	59,907.19
				Total	335,748.56

Table S9 (e) Material value from NMC battery cells

Pack material recycling

Recycled metal	Amount in waste stream (kg)	% recovery	Actual Recovery (kg)	Material price $/kg	Total recovered value ($)
Steel	6,305.80	0.52	3,279.02	0.67	2,201.31
Aluminum	18,347.70	0.6	11,008.62	2.08	22,862.22
Copper	3,979.93	0.53	2,109.36	7.33	15,453.14
Gold	0.87	0.96	0.83	45645.16	38,079.11
Palladium	0.24	0.7	0.17	23548.39	3,979.03
				Total	82,574.81

Table S9 (f) Material value from NMC battery pack materials

S10. Additional material input for cascaded use

The additional material input for assembly of a single refurbished EV LIB based stationary energy storage system (450 kWh) was obtained from Richa et al. (2015):

Additional input per stationary energy storage system	Mass (kg)
Copper connector	56.24
Battery management system (BMS)	56.02
Battery cabinet (steel)	55.25

Table S10 (a). Additional material input for assembly of a single stationary energy system

Mass composition of BMS (Richa et al., 2015):

BMS metal or component	Mass composition
Steel	41%
Aluminum	2%
Copper	5%
PWB	44%
Plastic	8%

Table S10 (b). Mass composition of BMS

S11. Cascaded Use Life Cycle Assessment

For the lead acid (PbA) battery system, the life cycle stages considered were battery production and operational charge-discharge energy efficiency losses (Rydh and Sanden 2005; Richa et al., 2015). For the refurbished EV LIB based stationary energy storage system, the life cycle stages that were included were battery refurbishment (transport of retired EV LIBs to refurbishment facility, cell testing and input of additional materials) and charge-discharge losses. LCI data for modeling these life cycle stages was obtained from life cycle assessment (LCA) study by Richa et al. (2015).

Based on the LCA data, the following results were obtained:

Stationary Energy Storage System	**MJ/kWh**	**CTUe/kWh**
Refurbished EV LIB	3321	211
Lead Acid Battery	4651	838

Table S11. Environmental impact of stationary energy storage system

S12. Comparison with BEV vehicle and battery production

The CED impact for producing a single vehicle was considered to be 88,400 MJ (Notter et al., 2010). The CED of LIB production was considered to be 949 MJ/kWh based on LCA data from Richa et al. (2015)[2]. Hence, for a 39 kWh BEV battery, the impact was 37,000 MJ/pack. The

[2] The eco-toxicity impact for LIB production was 222.85 CTUe/kWh (based on Richa et al., 2015).

total impact of manufacturing vehicle and battery was, thus 125,400 MJ. The avoided benefit per BEV pack from the case study was estimated to be 3,227 and 51,870 MJ for the reuse (in EV) and cascaded use application (stationary), respectively. The overall CED benefit/kg for the recycling pathway was estimated to be 24 MJ/kg. Considering the weight of a BEV battery (Nelson et al., 2011) as 330.5 kg, the benefit from recycling was estimated at 7,900 MJ/pack.

S13. Landfill Leaching Analysis

Sample Preparation

LIBs vary in composition, but are typically distinguished by cathode chemistry. The cathode chemistry selected for Toxicity Characteristic Leaching Procedure (TCLP) was $LiAlNiCoMnO_2$ and Panasonic LIB cells were used. The average composition of these cells adjusted for 50 g is shown below:

Material	Mass (g)	Mass fraction
Al	11.36	0.2272
Co	4.22	0.0845
Cu	8.30	0.1660
Fe	4.39	0.0879
Li	0.64	0.0128
Mn	2.93	0.0586
Ni	7.42	0.1484
Plastic	1.65	0.0329
Si	0.01	0.0002
P	1.02	0.0204
Zr	0.03	0.0006
O	2.26	0.0452
Binder	0.69	0.0139
C black	1.73	0.0347
S	0.01	0.0003
F	2.50	0.0499
Organic Solvent	0.83	0.0166
Total	50	1

Table S13 (a). Average composition of $LiAlNiCoMnO_2$ cells, adjusted for 50 g of cell mass

The batteries were treated by electrically discharging the cells and submerging them in liquid nitrogen for 5 minutes. Reduction in battery particle size was accomplished by shredding the batteries in an EconoGrind ESL180/180 Granulator for approximately five minutes. The battery material was then transferred to a fume hood to evaporate residual electrolyte and prepare them for additional physical processing. Four sieves of decreasing screen size (6 mm, 2.5 mm, 1.5 mm, 0.5 mm) were used to sort the battery material into manageable fractions and ensure that particle size <9.5 cm, per TCLP requirements.

Extraction fluid for the TCLP tests were selected by weighing out 5 g of battery material and combining it with 96.5 mL of distilled water in a 500 mL beaker. The beaker was covered with watch glass and stirred vigorously for 5 minutes. After the time had elapsed, the pH was measured to be >5. Using a pipette, 3.5 mL of 1 N HCl was added, slurried briefly and heated the solution to 50°C for 10 minutes. The solution was cooled to room temperature and the pH was measured to be >5, so Extraction Fluid 2 was selected.

TCLP Experiment

On an analytical balance, 100 g of battery material was weighed out and placed in a plastic bottle extractor. Two liters of Extraction Fluid 2 were prepared and added to the plastic bottle extractor by adding 5.7 mL of glacial acetic acid to a 1 L volumetric flask that was diluted to the 1 L mark with distilled water. The bottle extractor containing extraction fluid and battery material was placed on an 8-vessel rotary agitation apparatus (Analytical Testing Corporation) and agitated at 30 ± 2 RPM for 18 ± 2 hours. Once the time had elapsed, the solution was filtered using a pressure vessel and filter holder-containing a borosilicate glass fiber filter. The filtered solution was preserved by decreasing the pH to <2 with nitric acid and refrigerated until it could be analyzed. The experiment was completed in triplicate for each cell sample. The leachates were analyzed for potentially recyclable metals in LIBs (Al, Co, Cu, Fe, Li, Mn, Ni) using a Perkin Elmer Optima 8000 Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES).

Metal	leachate (mg/l)	Weight in 50 g sample (gm)	% leached (gm/gm)
Al	130.56	11.36	1.15%
Co	152.47	4.22	3.61%
Cu	1.61	8.30	0.02%
Fe	13.74	4.39	0.31%
Li	272.73	0.64	42.50%
Mn	335.27	2.93	11.45%
Ni	159.42	7.42	2.15%

Table S13 (b) Average Leaching results for LiAlCoMnNiO2 Panasonic cells

S14.Environmental impact of landfill pathway

Upstream Impacts: These impacts are considered to be from waste transport to landfill and landfill operation. For LIB waste from transport, an assumption of 0.5 tonne km/kg has been made based on Hischier et al. (2007). The CED and eco-toxicity impact data was obtained from SimaPro LCA software using ecoinvent database (ecoinvent Centre, 2010)

	MJ/kg	CTUe/kg
LIB waste transport to landfill	1.18	0.123
Landfill operation.	0.046	0.00108

Table S14 (a) Upstream CED and eco-toxicity impact data per kg of LIB landfill waste

Direct Toxicity Impacts: Eco-toxicity implications of landfill leaching of metals from the analyzed landfill was also estimated. Currently the eco-toxicity impacts of copper, manganese, iron, cobalt and nickel are characterized by USEtox LCA method.

LIB metal	USEtox Freshwater Ecotoxicity Characterization Factor (CTUe/kg)
Copper	2.62*10^6
Manganese	4.61*10^3
Iron	3.69*10^3
Cobalt	3.26*10^4
Nickel	7.73*10^4

Table S14 (b) USEtox Freshwater Ecotoxicity Characterization Factors for LIB metals

The landfill leaching potential of the EV LIB waste stream, under different scenarios of battery chemistry is presented below:

	LMO	NCM	LFP	Equal Mix
Iron leaching (kg)	9.77	9.47	31.78	17
Copper leaching (kg)	1.92	1.77	2.42	2
Manganese leaching (kg)	1,567.59	352.41		640
Cobalt leaching (kg)		81.19		27
Nickel leaching (kg)		64.66		22
Eco-toxicity impact of landfill leaching (CTUe)	12,281,585.68	13,937,965.19	6,456,008.36	10,891,853

Table S14 (c) Landfill leaching potential of the EV LIB waste stream, under different scenarios of battery chemistry

S15. Bill of materials of Lithium nickel manganese cobalt oxide (NMC) cells and pack

	Bill of materials of single cell (g)		
Materials	**BEV LIB cell**	**PHEV-12 LIB cell**	**PHEV-50 LIB cell**
Aluminum	62.53	150.38	58.20
Cobalt	46.14	40.17	42.58
Copper	82.59	246.16	77.12
Lithium	18.02	15.69	16.63
Manganese	43.01	37.45	39.70
Nickel	45.95	40.01	42.41
Graphite	134.91	117.11	124.63
Carbon	15.65	13.63	14.45
Binder	20.14	17.52	18.60
Plastic	15.83	44.67	14.68
Electrolyte	96.93	123.88	89.52
Other	79.07	68.84	72.98
No. of cells per pack	320	40	160

Table S15 (a)Bill of materials of Lithium nickel manganese cobalt oxide (NMC) cells (Based on Nelson et al., 2011)

Amount of materials in NMC cells (For single pack, in kg)			
Materials	BEV	PHEV-12	PHEV-50
Aluminum	20.01	6.02	9.31
Cobalt	14.76	1.61	6.81
Copper	26.43	9.85	12.34
Lithium	5.77	0.63	2.66
Manganese	13.76	1.50	6.35
Nickel	14.70	1.60	6.79
Graphite	43.17	4.68	19.94
Carbon	5.01	0.55	2.31
Binder	6.45	0.70	2.98
Plastic	5.07	1.79	2.35
Electrolyte	31.02	4.96	14.32
Other	25.30	2.75	11.68
Remaining NMC pack materials (For single pack, in kg)			
Materials	BEV	PHEV-12	PHEV-50
Steel	6.86	4.76	4.10
Aluminium	35.02	6.91	18.70
Copper	2.27	3.16	2.17
PWB	2.31	1.82	2.03
Plastic	0.44	0.35	0.39
Fiber glass	13.61	1.93	7.34
Coolant (Water+EC)	6.06	1.30	3.39
Total pack weight (kg)	**278.02**	**56.86**	**135.97**

Table S15 (b). Bill of materials of Lithium nickel manganese cobalt oxide (NMC) EV packs (Based on Nelson et al., 2011)

S16. Bill of materials of Lithium Ferrous Phosphate (LFP) cells and pack

Bill of materials of single LFP cell (g)			
	BEV	PHEV-12	PHEV-50
Aluminum	100.18	137.87	93.08
Copper	136.60	209.81	127.23
Lithium	11.17	9.96	10.31
Iron	89.93	80.18	83.00
Graphite	141.06	125.73	130.29
Carbon	17.12	15.26	15.80
Binder	21.69	19.34	20.02
Plastic	26.62	39.31	24.65
Electrolyte	184.29	184.24	170.15
Other	152.84	136.28	141.07
Total per cell (g)	881.49	957.98	815.61

Table S16 (a). Bill of materials of LFP cells (Based on Nelson et al., 2011)

For modeling the EV reuse scenario for a waste stream comprised entirely of LFP cathode based EV LIB packs: The environmental impact for LIB pack manufacturing (for estimating avoided impact of producing new replacement batteries for EV application) and cell manufacturing (for estimating the refurbishment impact for replacing 10% cells in an old LIB) was calculated from LCI data collected from battery bill of materials, Richa et al. (2015**),** Majeau-Bettez et al. (2011), and Notter et al. (2010). The battery pack was modeled in Simapro LCA software. The following results were obtained for a 369 kg BEV pack, storing 39KWh energy with a specific energy of 106 Wh/kg:

	MJ/kg	CTUe/kg
LFP cells	91.6	24.4
Pack components and pack assembly	41.4	12.2
LFP pack	79.7	21.5

Table S16 (b). LCA data for LFP battery pack

Amount of materials in LFP cells (For single pack, in kg)			
Materials	BEV	PHEV-12	PHEV-50
Aluminum	32.06	5.51	14.89
Copper	43.71	8.39	20.36
Lithium	3.58	0.40	1.65
Iron	28.78	3.21	13.28
Graphite	45.14	5.03	20.85
Carbon	5.48	0.61	2.53
Binder	6.94	0.77	3.20
Plastic	8.52	1.57	3.94
Electrolyte	58.97	7.37	27.22
Other	48.91	5.45	22.57
Remaining LFP pack materials (For single pack, in kg)			
Materials	BEV	PHEV-12	PHEV-50
Steel	9.07	5.30	5.11
Aluminium	48.24	9.00	25.65
Copper	2.65	3.41	2.35
PWB	2.31	1.82	2.03
Plastic	0.44	0.35	0.39
Fiber glass	17.04	3.21	9.11
Coolant (Water+EC)	7.50	1.45	3.77
Total per pack (kg)	**369.33**	**62.87**	**178.90**

Table S16(c). Bill of materials of LFP EV packs (Based on Nelson et al., 2011)

S17. Contributions to net results (environmental impacts)

1. Reuse in EVs

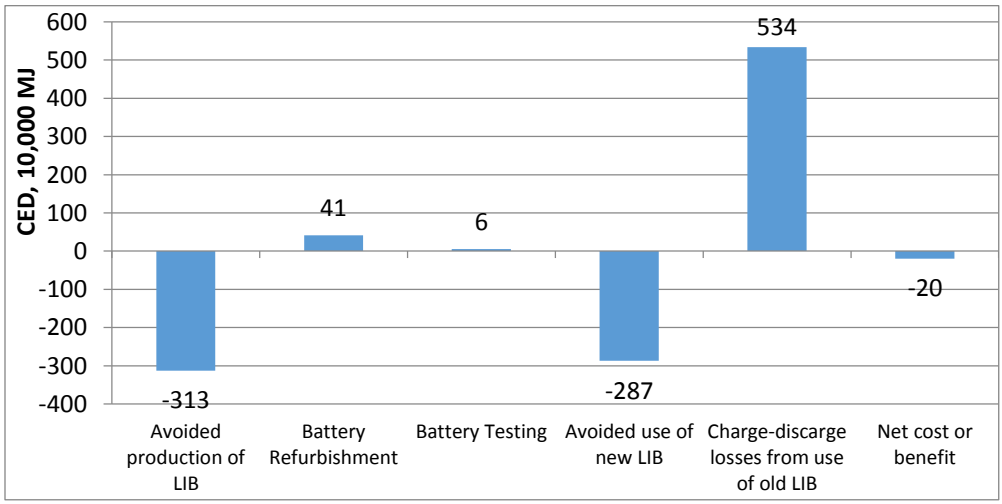


Fig S17 (a).Contributors to net CED for EV reuse (Net avoided CED=200,000 MJ)

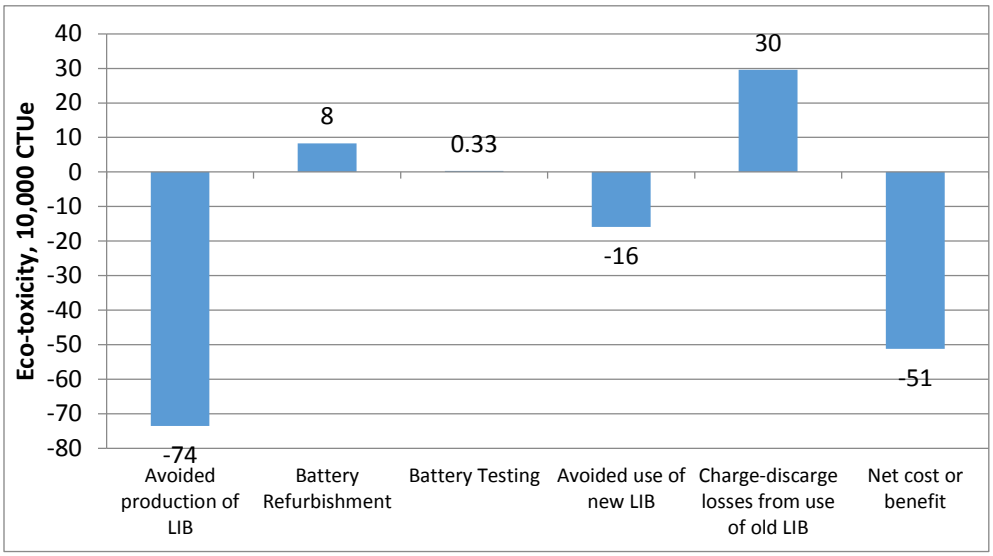


Fig S17 (b).Contributors to net eco-toxicity for EV reuse (Net avoided eco-toxicity=500,000 CTUe)

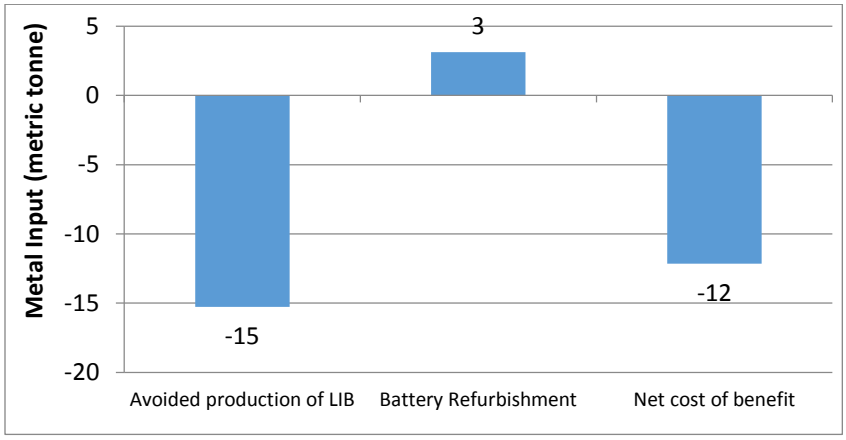


Fig S17(c).Contributors to net metal input for EV reuse (Net avoided metal input=12 mT)

2. Cascaded Use

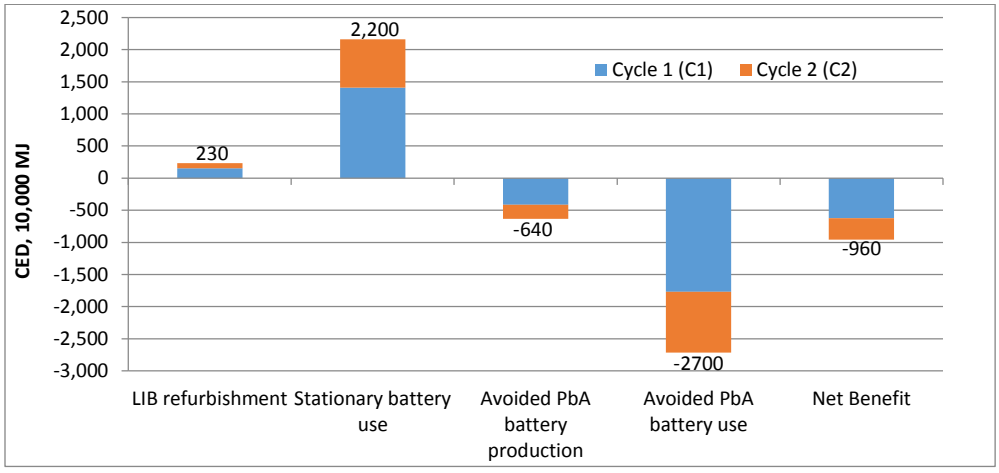


Fig S17 (d).Contributors to net CED for cascaded use (Net avoided CED=9.6 million MJ)

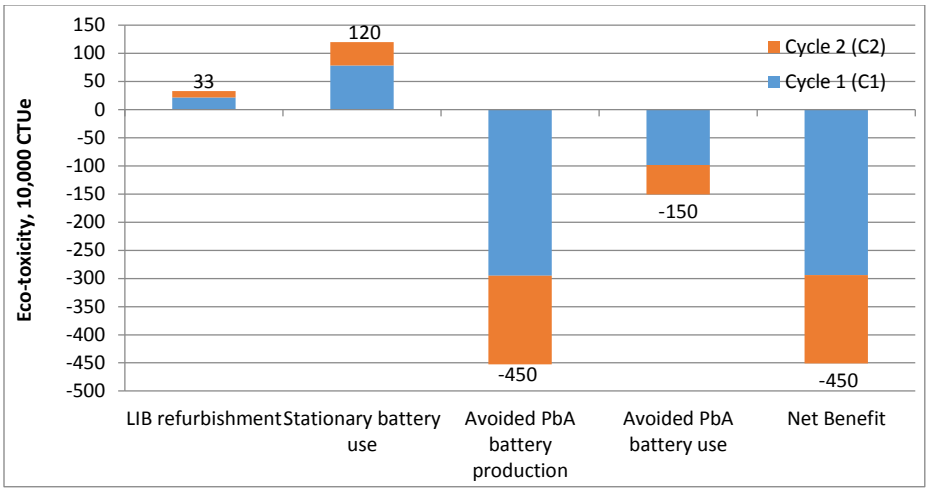


Fig S17 (e).Contributors to net eco-toxicity for cascaded use (Net avoided eco-toxicity=4.5 million CTUe)

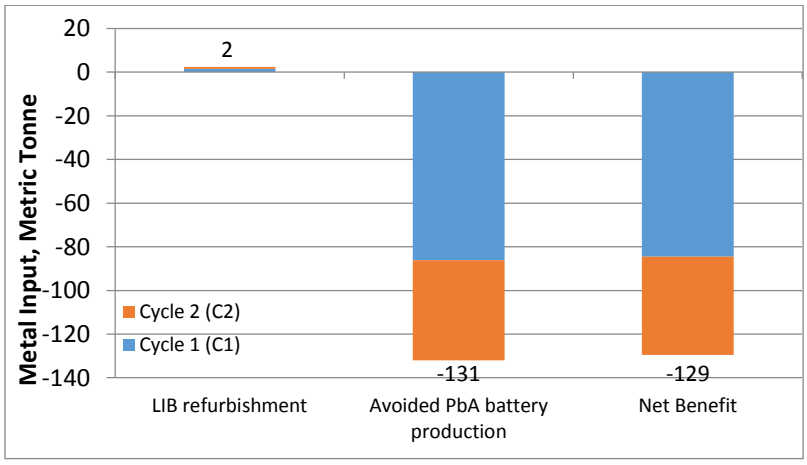


Fig S17 (f).Contributors to net metal input for cascaded use (Net avoided metal input=130 mT)

3. Recycling

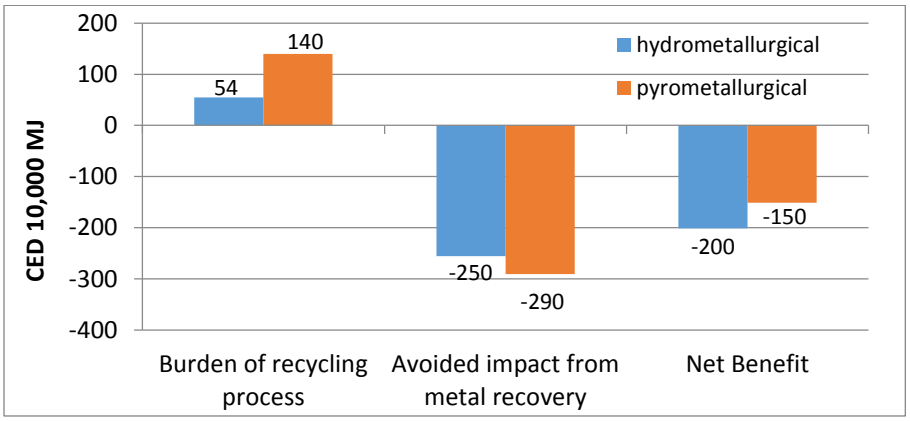


Fig S17 (f).Contributors to net CED for recycling (Net avoided CED=3.5 million MJ)

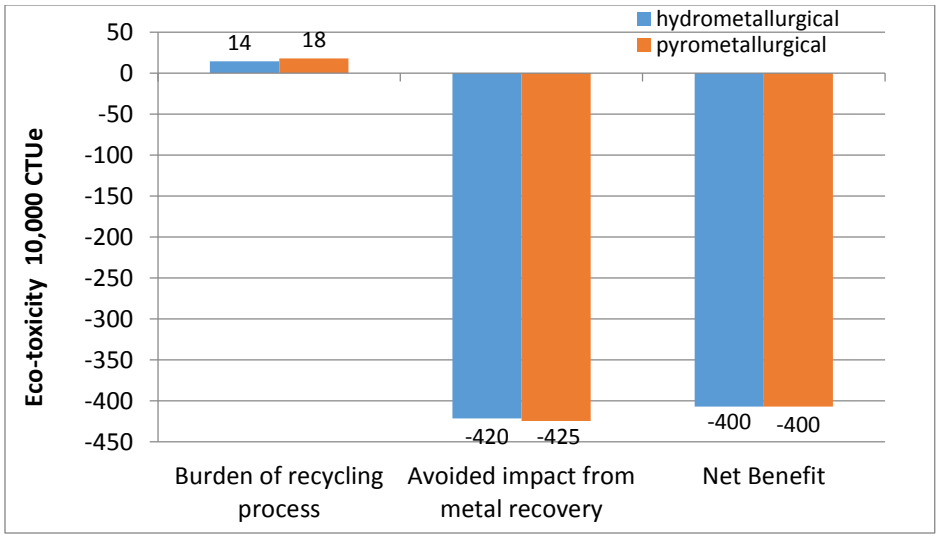


Fig S17 (g).Contributors to net eco-toxicity for recycling (Net avoided eco-toxicity=8 million CTUe)

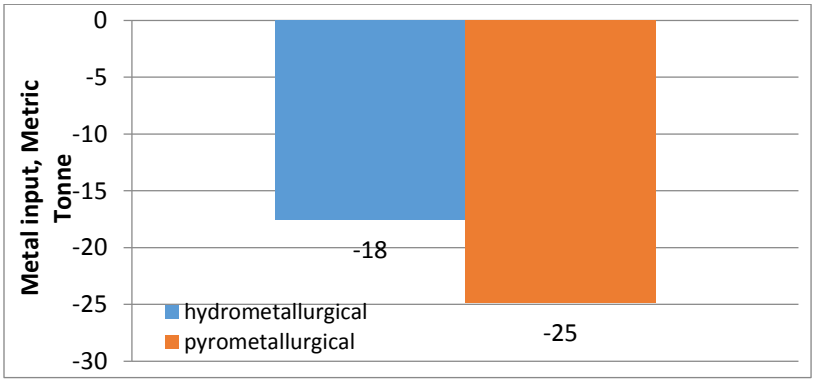


Fig S17 (h).Contributors to net avoided metal input from recycling (Net avoided metal input = 43 mT)

S18. Drinking water standards

Metal	Average leachate concentration (mg/L)	US Primary standard (mg/L)	US Secondary Standard (mg/L)	EU directive (mg/L)	World Health Organization (mg/L)
Aluminum	130.5		0.05-0.2	0.2	0.1-0.2
Copper	1.61	1.3	1	2	2
Iron	13.74		0.3	0.2	
Manganese	335.27		0.05	0.05	0.4
Nickel	159.42			0.02	0.07

Table S18. National and international drinking water regulations for LIB metals, in mg/L.
Data Source: US EPA (2009), European Union (1998), and World Health Organization (2008)

REFERENCES

1. Ahmadi, L., Fowler, M., Young, S. B., Fraser, R. A., Gaffney, B., & Walker, S. B. (2014b). Energy efficiency of Li-ion battery packs re-used in stationary power applications. *Sustainable Energy Technologies and Assessments*, *8*, 9-17.
2. Ahmadi, L., Yip, A., Fowler, M., Young, S. B., & Fraser, R. A. (2014a). Environmental feasibility of re-use of electric vehicle batteries. *Sustainable Energy Technologies and Assessments*, *6*, 64-74.
3. Alber, G. (n.d.) Predicting battery performance using internal cell resistance. http://www.alber.com/docs/predictbatt.pdf. Accessed 13 November 2014
4. Albercorp. (n.d.) Internal resistance and the cellcorder. http://alber.com/Docs/CC1Ban.pdf. Accessed 13 November 2014
5. Albright, G., Edie, J., & Al-Hallaj, S. (2012). A Comparison of Lead-acid to Lithium-ion in Stationary Storage Applications. AllCell Technologies LLC. http://www.batterypoweronline.com/main/wp-content/uploads/2012/07/Lead-acid-white-paper.pdf. Accessed 20 October 2014
6. Allwood, J. M., Ashby, M. F., Gutowski, T. G., & Worrell, E. (2011). Material efficiency: A white paper. *Resources, Conservation and Recycling*, *55*(3), 362-381.
7. Allwood, J.M., Cullen, J.M., Carruth, M.A., Cooper, D.R., McBrien, M., Milford, R.L., Moynihan, M.C. and Patel, A.C. (2012). *Sustainable materials: with both eyes open* (p. 64). Cambridge: UIT Cambridge.
8. Alonso, E., Sherman, A. M., Wallington, T. J., Everson, M. P., Field, F. R., Roth, R., & Kirchain, R. E. (2012). Evaluating rare earth element availability: a case with revolutionary demand from clean technologies. *Environmental science & technology*, *46*(6), 3406-3414.
9. Amarakoon, S., Smith, J., & Segal, B. (2013). *Application of life-cycle assessment to nanoscale technology: Lithium-ion batteries for electric vehicles* (No. EPA 744-R-12-001).
10. Anderman, M. (2007). The 2007 Advanced Automotive Battery and Ultracapacitor Industry Report. *Advanced Automotive Batteries*.
11. Andersson, A. M., Abraham, D. P., Haasch, R., MacLaren, S., Liu, J., & Amine, K. (2002). Surface characterization of electrodes from high power lithium-ion batteries. *Journal of The Electrochemical Society*, *149*(10), A1358-A1369.
12. Arbabzadeh, M., Johnson, J. X., Keoleian, G. A., Rasmussen, P. G., & Thompson, L. T. (2015). Twelve Principles for Green Energy Storage in Grid Applications. *Environmental science & technology*, *50*(2), 1046-1055.
13. Archuleta, M. M. (1995). Toxicity of materials used in the manufacture of lithium batteries. *Journal of power sources*, *54*(1), 138-142.
14. Argonne National Laboratory [ANL]. (2011). BatPac (Battery Performance and Cost Model) Version 2.0. http://www.cse.anl.gov/batpac/. Accessed 21 December 2013
15. Argonne National Laboratory. (2012). BatPaC Model. Retrieved from http://www.cse.anl.gov/batpac/
16. Arora, P., White, R. E., & Doyle, M. (1998). Capacity fade mechanisms and side reactions in lithium-ion batteries. *Journal of the Electrochemical Society*, *145*(10), 3647-3667.
17. Arora, P., & Zhang, Z. (2004). Battery separators. *Chemical reviews*, *104*(10), 4419-4462.
18. Asari, M., & Sakai, S. I. (2013). Li-ion battery recycling and cobalt flow analysis in Japan. *Resources, Conservation and Recycling*, *81*, 52-59.

19. Axsen, J., Kurani, K. S., & Burke, A. (2010). Are batteries ready for plug-in hybrid buyers? *Transport Policy*, *17*(3), 173-182.
20. Axsen, J., Burke, A. F., & Kurani, K. S. (2008). *Batteries for plug-in hybrid electric Vehicles (PHEVs): goals and the state of technology circa 2008*. Institute of Transportation Studies, University of California, Davis.
21. Azapagic, A., & Clift, R. (1999). Allocation of environmental burdens in multiple-function systems. *Journal of Cleaner Production*, *7*(2), 101-119.
22. Babbitt, C. W., Williams, E., & Kahhat, R. (2011). Institutional disposition and management of end-of-life electronics. *Environmental science & technology*, *45*(12), 5366-5372.
23. Bakker, C., Wang, F., Huisman, J., & den Hollander, M. (2014). Products that go round: exploring product life extension through design. *Journal of Cleaner Production*, *69*, 10-16.
24. Batayneh, M., Marie, I., & Asi, I. (2007). Use of selected waste materials in concrete mixes. *Waste management*, *27*(12), 1870-1876.
25. Battery Council International. (2012). Battery Recycling. *Battery Council International*. http://batterycouncil.org/?page=battery_recycling. Accessed 21 December 2013
26. Beer, S., Gómez, T., Dallinger, D., Momber, I., Marnay, C., Stadler, M., & Lai, J. (2012). An economic analysis of used electric vehicle batteries integrated into commercial building microgrids. *IEEE Transactions on Smart Grid*, *3*(1), 517-525.
27. Berdichevsky, G., Kelty, K., Straubel, J. B., & Toomre, E. (August, 2006). The Tesla Roadster battery system. *Tesla Motors*. Retrieved from http://webarchive.teslamotors.com/display_data/TeslaRoadsterBatterySystem.pdf
28. Bindner, H., Cronin, T., Lundsager, P., Manwell, J. F., Abdulwahid, U., & Baring-Gould, I. (2005). *Lifetime modelling of lead acid batteries*. Risø National Laboratory Roskilde, Denmark. http://orbit.dtu.dk/fedora/objects/orbit:88309/datastreams/file_7710966/content. Accessed 20 October 2014
29. Blanco, S. (May, 2010). Details on Nissan Leaf battery pack, including how recharging speed affects battery life. AutoblogGreen, AOL Inc. http://green.autoblog.com/2010/05/27/details-on-nissan-leaf-battery-pack-including-how-recharging-sp/. Accessed 20 November 2014
30. Boden, D. P. (1998). Selection of pre-blended expanders for optimum lead/acid battery performance. *Journal of power sources*, *73*(1), 89-92.
31. Boguski, T. K., Hunt, R. G., & Franklin, W. E. (1994). General mathematical models for LCI recycling. *Resources, Conservation and Recycling*, *12*(3), 147-163.
32. Bond, V. (2013). 2nd lives for EV batteries could cut prices. *Automotive News*. http://www.autonews.com/article/20130506/OEM05/305069985/2nd-lives-for-ev-batteries-could-cut-prices. Accessed 19 January 2015
33. Borg, M., Paulsen, J., & Trinius, W. (2001). Proposal of a method for allocation in building-related environmental LCA based on economic parameters. *The international journal of life cycle assessment*, *6*(4), 219-230.
34. Borken-Kleefeld, J., & Weidema, B. P. (2013). Global default data for freight transport per product group. *Manuscript for special ecoinvent 3.0 issue of the International Journal of Life Cycle Assessment.*
35. Brandstotter, M., Knoth, R., Kopacek, B., & Kopacek, P. (2004). Case study of a printed-wire-board concerning (re-) design for environment. *IEEE transactions on electronics packaging manufacturing*, *27*(1), 26-32.

36. Brunner, P. H., & Rechberger, H. (2004). Practical handbook of material flow analysis. *New York: Lewis*, *317*.
37. Burke, A. (2009). *Performance, charging, and second-use considerations for lithium batteries for plug-in electric vehicles*. Institute of Transportation Studies, University of California-Davis. Retrieved from http://escholarship.org/uc/item/2xf263qp#page-1
38. Burke, A. F., & Miller, M. (2009). *Performance characteristics of lithium-ion batteries of various chemistries for plug-in hybrid vehicles*. Institute of Transportation Studies, University of California, Davis, Research Report UCD-ITS-RR-09-08. Retrieved from http://publications.its.ucdavis.edu/publication_detail.php?id=1292
39. California Code [CA Code]. (2006). Rechargeable Battery Recycling Act of 2006 (42451.e 42456.) (Chapter 8.4)
40. Call2Recycle. (2015). What can I recycle? www.call2recycle.org/what-can-i-recycle/. Accessed January 2015.
41. Campanari, S., Manzolini, G., & Garcia de la Iglesia, F. (2009). Energy analysis of electric vehicles using batteries or fuel cells through well-to-wheel driving cycle simulations. *Journal of Power Sources*, *186*(2), 464-477.
42. Celik, A. N., Muneer, T., & Clarke, P. (2008). Optimal sizing and life cycle assessment of residential photovoltaic energy systems with battery storage. *Progress in Photovoltaics: Research and Applications*, *16*(1), 69-85.
43. Chalk, S. G., & Miller, J. F. (2006). Key challenges and recent progress in batteries, fuel cells, and hydrogen storage for clean energy systems. *Journal of Power Sources*, *159*(1), 73-80
44. Chang, T., You, S. , Yu, B. , & Yao, K. (2009). A material flow of lithium batteries in Taiwan. *Journal of Hazardous Materials*, *163*(2-3), 910-915
45. Chen, L.; Tang, X.; Zhang, Y.; Li, L.; Zeng, Z.; Zhang, Y. (2011). Process for the recovery of cobalt oxalate from spent lithium-ion batteries. *Hydrometallurgy*, *108* (1-2), 80-86.
46. Chiang, Y. M. (April 11, 2013). Advances in lithium ion technology and applications. National Transportation Safety Board, Li-ion Battery Forum, April 2013, Washington D.C. Retrieved from http://www.ntsb.gov/news/events/2013/batteryforum/presentations/Chiang%20Presentation%20-%20Battery%20Forum.pdf
47. Chiodo, J. (2005). Design for disassembly guidelines. *Active Disassembly Research*, *2*(1), 29-37.
48. Choi, D., Wang, W., Viswanathan, V. V., & Yang, G. Z. (2010). Low Cost, Long Cycle Life, Li-ion Batteries for Stationary Applications. *DOE EERE Energy Storage Program Annual Update 2010*, Washington DC
49. Cicconi, P., Landi, D., Morbidoni, A., & Germani, M. (2012, September). Feasibility analysis of second life applications for Li-ion cells used in electric powertrain using environmental indicators. In *Energy Conference and Exhibition (ENERGYCON), 2012 IEEE International* (pp. 985-990). IEEE.
50. Cleary, J. (2009). Life cycle assessments of municipal solid waste management systems: a comparative analysis of selected peer-reviewed literature. *Environment International*, *35*(8), 1256-1266.
51. Cready, E., Lippert, J., Pihl, J., Weinstock, I., Symons, P., Jungst, R. G. (2003). *Technical and Economic Feasibility of Applying Used EV Batteries in Stationary Applications*. SAND2002-4084; Sandia National Laboratories: Albuquerque, NM.

52. Dahn, J., & Erlich, G. M. (2011). Lithium-ion Batteries. *Linden's Handbook of Batteries, 4th edition, TB Reddy, Ed*, 26.
53. Davis E., Funk, D., & Johnson, W. (2002). Internal Ohmic Measurements and Their Relationship to Battery Capacity–EPRI's Ongoing Technology Evaluation. *In Proceedings of the 2002 BATTCON Conference, Ft. Lauderdale, FL*
54. Davis, S. C., Diegel, S. W., & Boundy, R. G. (2013). *Transportation energy data book: Edition 32* (No. ORNL 6989). United States. Department of Energy. Retrieved from http://cta.ornl.gov/data/tedb32/Edition32_Full_Doc.pdf
55. Denholm, P. & Short, W. (2006). An evaluation of utility system impacts and benefits of optimally dispatched plug-in hybrid electric vehicles. Technical Report NREL/TP-620-40293, National Renewable Energy Laboratory, Golden, Colorado.
56. Dewulf, J., Van der Vorst, G., Denturck, K., Van Langenhove, H., Ghyoot, W., Tytgat, J., & Vandeputte, K. (2010). Recycling rechargeable lithium ion batteries: critical analysis of natural resource savings. *Resources, Conservation and Recycling*, *54*(4), 229-234.
57. Dinger, A., Martin, R., Mosquet, X., Rabl, M., Rizoulis, D., Russo, M., & Sticher, G. (2010). Batteries for Electric Cars: Challenges, Opportunities, and the Outlook to 2020. *The Boston Consulting Group, Technical Report*. Retrieved from http://www.bcg.com/documents/file36615.pdf
58. Doggett, S. (2011). GM and ABB give Chevy Volt Battery second life. Edmunds Auto Observer. http://www.edmunds.com/autoobserver-archive/2011/07/gm-and-abb-give-chevy-volt-battery-second-life.html. Accessed 12 April 2013
59. Dorella, G.; Mansur, M. B. (2007). A study of the separation of cobalt from spent Li-ion battery residues. *Journal of Power Sources*, *170* (1), 210-215.
60. Doughty, D. H., Butler, P. C., Akhil, A. A., Clark, N. H., & Boyes, J. D. (2010). Batteries for large-scale stationary electrical energy storage. *The Electrochemical Society Interface*, *19*(3), 49-53.
61. Dunn, B., Kamath, H., & Tarascon, J. M. (2011). Electrical energy storage for the grid: a battery of choices. *Science*, *334*(6058), 928-935.
62. Dunn, J. B., Gaines, L., Barnes, M., Sullivan, J., & Wang, M. Q. (2012). *Material and energy flows in materials production, assembly, and end-of-life stages of the life cycle of lithium-ion batteries*; ANL/ESD/12-3; Argonne National Laboratory.
63. Dunn, J. B., Gaines, L., Kelly, J. C., James, C., & Gallagher, K. G. (2015). The significance of Li-ion batteries in electric vehicle life-cycle energy and emissions and recycling's role in its reduction. *Energy & Environmental Science*, *8*(1), 158-168.
64. Duvall, M. & Knipping, E. (2007). Environmental Assessment of Plug-In Hybrid Electric Vehicles, Volume 1: Nationwide Greenhouse Gas Emissions. EPRI, Palo Alto, CA. Report #1015325. Retrieved from http://et.epri.com/publicdocuments.html.
65. Eberhard, M., & Tarpenning, M. (October, 2006). *The 21st Century Electric Car*. Tesla Motors Inc. Retrieved from http://www.stanford.edu/group/greendorm/participate/cee124/TeslaReading.pdf
66. ecoinvent Centre. (2007). ecoinvent data v2.0. ecoinvent reports No.1-25, Swiss Centre for Life Cycle Inventories, Dübendorf, 2007.
67. ecoinvent Centre. (2010). ecoinvent data and reports v.2.2. Dübendorf, Switzerland: Swiss Center for Life Cycle Inventories
68. EIA. (2012). Annual Energy Outlook 2012. *Light-duty vehicle sales by technology - United States*. http://www.eia.gov/forecasts/archive/aeo12/

69. Ekvall, T., & Tillman, A. M. (1997). Open-loop recycling: criteria for allocation procedures. *The international journal of life cycle assessment*, *2*(3), 155-162.
70. Elkind, E.N. (2014). Reuse and Repower-How to save money and clean the grid with second-life electric vehicle batteries. www.law.berkeley.edu/files/ccelp/Reuse_and_Repower_--_Web_Copy.pdf. Accessed 21 January 2015
71. Ellen MacArthur Foundation. (2013). Towards the circular economy. Ellen MacArthur Foundation. Retrieved from http://mvonederland.nl/system/files/media/towards-the-circular-economy.pdf
72. Ellingsen, L. A. W., Majeau-Bettez, G., Singh, B., Srivastava, A. K., Valøen, L. O., & Strømman, A. H. (2014). Life cycle assessment of a lithium-ion battery vehicle pack. *Journal of Industrial Ecology*, *18*(1), 113-124.
73. EPA . (n.d.). Hydrogen fluoride study. Final report to Congress: Section 112(n) (6) Clean Air Act as amended. Retrieved from http://www.epa.gov/osweroe1/docs/chem/hydro.pdf
74. Espinosa, D. C. R., Bernardes, A. M., & Tenório, J. A. S. (2004). An overview on the current processes for the recycling of batteries. *Journal of Power Sources*, *135*(1), 311-319.
75. Espinoza, V. S., Erbis, S., Pourzahedi, L., Eckelman, M. J., & Isaacs, J. A. (2014). Material flow analysis of carbon nanotube lithium-ion batteries used in portable computers. *ACS Sustainable Chemistry & Engineering*, *2*(7), 1642-1648.
76. EU. (2000). Directive 2000/53EC of the European Parliament and of the Council (2000.9.18)
77. Eurofins. (2012). Federal and State Hazardous Waste Criteria. http://www.eurofinsus.com/media/161417/hazardous_waste_regulatory_limits.pdf
78. European Parliament. (2008). European Parliament Council directive on waste and repealing certain directives. 2008/98/EC. *Official Journal* L 312 (2008), pp. 3–30.
79. European Commission. (2014). Development of Guidance on Extended Producer Responsibility (EPR). Final Report. http://ec.europa.eu/environment/waste/pdf/target_review/Guidance%20on%20EPR%20-%20Final%20Report.pdf. Accessed 27 January 2015
80. European Commission. (2006). Directive 2006/66/EC of the European Parliament and the Council of 6 September 2006 on batteries and accumulators and waste batteries and accumulators and repealing Directive 91/157/EEC. *Official Journal of the European Communities* 1e14: 2006a.
81. European Telecommunications Standards Institute [ETSI]. (2012). Environmental Engineering (EE); The use of alternative energy solutions in telecommunication installations. Technical Report. ETSI TR 102 532 V1.2.1 (2012-11). http://www.etsi.org/deliver/etsi_tr/102500_102599/102532/01.02.01_60/tr_102532v010201p.pdf. Accessed 25 December 2013
82. European Union. (1998). Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption. http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:31998L0083&from=EN. Accessed December 2015.
83. Fenton, M. D. (2003). *Iron and steel recycling in the United States in 1998*. US Department of the Interior, U.S. Geological Survey.
84. Ferreira, D. A., Prados, L. M. Z., Majuste, D., & Mansur, M. B. (2009). Hydrometallurgical separation of aluminium, cobalt, copper and lithium from spent Li-ion batteries. *Journal of Power Sources*, *187* (1), 238-246.

85. Fisher, K., Wallén, E., Laenen, P. P., & Collins, M. (2006). Battery waste management life cycle assessment. *UK Department for Environment Food and Rural Affairs (Defra), UK*.
86. Frampton, J. (1998). Comparison of California's Waste Extraction Test (WET) and the U.S. EPA's Toxicity Characteristic Procedure (TCLP). California Department of Toxic Substances Control.
87. Frost & Sullivan. (2010). Global Electric Vehicles Lithium-ion Battery Second Life and Recycling Market Analysis, M5B6-18, November 2010.
88. Frost & Sullivan. (2009). World Hybrid Electric Vehicle/Plug-in Hybrid Electric Vehicle Battery Market, N627- 27, September 2009.
89. Frost & Sullivan. (2014). 2020 Vision: Global Lithium-Ion Battery Market. Frost & Sullivan, San Antonio.
90. Fu, H. (2009). *Implementations of electric vehicle system based on solar energy in Singapore assessment of lithium-ion batteries for automobiles* (Doctoral dissertation, Massachusetts Institute of Technology). Retrieved from http://dspace.mit.edu/handle/1721.1/54200
91. Gaines, L. (2014). The future of automotive lithium-ion battery recycling: Charting a sustainable course. *Sustainable Materials and Technologies*, *1*, 2-7.
92. Gaines, L., & Nelson, P. (2010). *Lithium-ion batteries: examining material demand and recycling issues*. Argonne, IL: Argonne National Laboratory.
93. Gaines, L., & Cuenca, R. (2000). Costs of Lithium-Ion Batteries for Vehicles. Argonne National Laboratories. Retrieved from http://ntl.bts.gov/lib/14000/14700/14729/DE00761281.pdf
94. Ganter, M. J., Landi, B. J., Babbitt, C. W., Anctil, A., & Gaustad, G. (2014). Cathode refunctionalization as a lithium ion battery recycling alternative. *Journal of Power Sources*, *256*, 274-280.
95. Garetson, T. (2013). Advanced Vehicle Testing and Evaluation. Final Report. US Department of Energy Vehicle Technologies Office. Retrieved from http://www.osti.gov/scitech/servlets/purl/1097102. Accessed 19 December 2014
96. Geiser, K. (2001). Materials matter: Toward a sustainable materials policy: MIT Press.
97. Geisler, G., Hofstetter, T. B., & Hungerbühler, K. (2004). Production of fine and speciality chemicals: procedure for the estimation of LCIs. *The International Journal of Life Cycle Assessment*, *9*(2), 101-113.
98. Genikomsakis, K. N., Ioakimidis, C. S., Murillo, A., Trifonova, A., & Simic, D. (2013, November). A Life Cycle Assessment of a Li-ion urban electric vehicle battery. In *Electric Vehicle Symposium and Exhibition (EVS27), 2013 World*(pp. 1-11). IEEE.
99. Georgi-Maschler, T., Friedrich, B., Weyhe, R., Heegn, H., & Rutz, M. (2012). Development of a recycling process for Li-ion batteries. *Journal of power sources*, *207*, 173-182.
100. Ghisellini, P., Cialani, C., & Ulgiati, S. (2016). A review on circular economy: The expected transition to a balanced interplay of environmental and economic systems. *Journal of Cleaner Production*, *114*, 11-32.
101. Giffi, C., Gardner, M., Hill, R., & Hasegawa, M. (January, 2010). Gaining Traction- A Customer View of Electric Vehicle Mass Adoption in the US Automotive Market. Deloitte Development LLC. Retrieved from http://www.deloitte.com.br/publicacoes/2007/MFG.Gaining_Traction_customer_view_of_electric_vehicle_mass_adoption.pdf
102. Glew, D., Stringer, L. C., & McQueen-Mason, S. (2013). Achieving sustainable biomaterials by maximising waste recovery. *Waste management*, *33*(6), 1499-1508.

103. Gondelach, S. (2010, November). *Current and future developments of batteries for electric cars-an analysis.* Retrieved from http://igitur-archive.library.uu.nl/student-theses/2010-1126-200244/UUindex.html
104. Gordon-Bloomfield, N. (2015). BMW, Bosch, Vattenfall Launch Second Life Batteries Alliance to Give Electric Car Batteries A Retirement Plan. Transport Evolved. https://transportevolved.com/2015/01/27/bmw-bosh-vattenfall-launch-second-life-batteries-alliance-give-electric-car-batteries-retirement-plan/. Accessed 28 January 2015
105. Graedel, T. E., & Allenby, B. R. (2010). *Industrial ecology and sustainable engineering*. Upper Saddle River, NJ, USA: Prentice Hall.
106. Graedel, T. E., Allwood, J., Birat, J. P., Buchert, M., Hagelüken, C., Reck, B. K., & Sonnemann, G. (2011). *Recycling rates of metals: A status report*. United Nations Environment Programme.
107. Greene, D. L., & DeCicco, J. (2000). Engineering-Economic Analyses of Automotive fuel Economy Potential in the United States 1. *Annual Review of Energy and the Environment*, 25(1), 477-535.
108. Greene, D. L., Patterson, P. D., Singh, M., & Li, J. (2005). Feebates, rebates and gas-guzzler taxes: a study of incentives for increased fuel economy. *Energy Policy*, 33(6), 757-775
109. Gregson, N., Crang, M., Fuller, S., & Holmes, H. (2015). Interrogating the circular economy: the moral economy of resource recovery in the EU. *Economy and Society*, *44*(2), 218-243.
110. Grosjean, C., Miranda, P. H., Perrin, M., & Poggi, P. (2012). Assessment of world lithium resources and consequences of their geographic distribution on the expected development of the electric vehicle industry. *Renewable and Sustainable Energy Reviews*, *16*(3), 1735-1744.
111. Gruber, P.W., Medina, P.A., Keoleian, G.A., Kesler, S.E., Everson, M.P., & Wallington, T.J. (2011). Global lithium availability a constraint for electric vehicles? *Journal of Industrial Ecology*, *15*(5), 760-775
112. Guinée, J. B., Heijungs, R., & Huppes, G. (2004). Economic allocation: examples and derived decision tree. *The international journal of life cycle assessment*, *9*(1), 23-33.
113. Gutowski, T. G., Sahni, S., Boustani, A., & Graves, S. C. (2011). Remanufacturing and energy savings. *Environmental science & technology*,*45*(10), 4540-4547.
114. Han, X., Ouyang, M., Lu, L., & Li, J. (2014). A comparative study of commercial lithium ion battery cycle life in electric vehicle: Capacity loss estimation. *Journal of Power Sources*, *268*, 658-669.
115. Harper, E. M., Kavlak, G., & Graedel, T. E. (2011). Tracking the Metal of the Goblins: Cobalt's Cycle of Use. *Environmental Science & Technology*, *46*(2), 1079-1086.
116. Hawkins, T. R., Singh, B., Majeau-Bettez, G., & Strømman, A. H. (2013). Comparative environmental life cycle assessment of conventional and electric vehicles. *Journal of Industrial Ecology*, *17*(1), 53-64.
117. Hein, R., Kleindorfer, P. R., & Spinler, S. (2012). Valuation of electric vehicle batteries in vehicle-to-grid and battery-to-grid systems. *Technological Forecasting and Social Change*, *79*(9), 1654-1671.
118. Hendrickson, T. P., Kavvada, O., Shah, N., Sathre, R., & Scown, C. D. (2015). Life-cycle implications and supply chain logistics of electric vehicle battery recycling in California. *Environmental Research Letters*, *10*(1), 014011.

119. Heymans, C., Walker, S. B., Young, S. B., & Fowler, M. (2014). Economic analysis of second use electric vehicle batteries for residential energy storage and load-levelling. *Energy Policy*, *71*, 22-30.
120. Hill, K., & Cregger, J. (2011). Deployment Rollout Estimate of Electric Vehicles, 2011 - 2015. Center for Automotive Research. January 2011. Retrieved from http://www.cargroup.org/assets/files/deployment.pdf
121. Hischier, R., Classen, M., Lehmann, M., & Scharnhorst, W. (2007). Life cycle inventories of electric and electronic equipment: production, use and disposal. *Final report ecoinvent Data v2. 0*, *18*.
122. Hittman Associates. (1980). Life cycle energy analysis of electric vehicle storage batteries, H-1008/001-80-964, submitted to the US Department of Energy, contract number DE-AC02-79ET25420.A000
123. Hoffart, F. (2008). Proper care extends Li-ion battery life. Power Electronics Technology Magazine. http://powerelectronics.com/. Accessed 26 November 2014
124. Howard, W., & Spotnitz, R. (2007). Theoretical evaluation of high-energy lithium metal phosphate cathode materials in li-ion batteries. *Journal of Power Sources*, 165(2), 887-891.
125. Howell, D. (2012). Overview of Battery R&D Activities. Washington DC: U.S. Department of Energy Vehicle Technologies Program. Retrieved from http://energy.gov/sites/prod/files/2014/03/f9/vtpn07_es_howell_2012_o.pdf. Accessed January 2016.
126. Hoyer, C., Kieckhäfer, K., & Spengler, T. S. (2011). A strategic framework for the design of recycling networks for lithium-ion batteries from electric vehicles. In *Glocalized Solutions for Sustainability in Manufacturing* (pp. 79-84). Springer Berlin Heidelberg.
127. Huang, S., Hodge, B. M. S., Taheripour, F., Pekny, J. F., Reklaitis, G. V., & Tyner, W. E. (2011). The effects of electricity pricing on PHEV competitiveness. *Energy Policy*, 39(3), 1552-1561.
128. Humphrey, J., Sargent, D., Schuster, J., Marshall, M., Omotoso, M., & Dunne, T. (2010). Drive Green 2020: More hope than reality. J.D. Power and Associates. Retrieved from http://www.altomelbilen.dk/presentationer/DriveGreen2020.pdf
129. Iakovou, E., Moussiopoulos, N., Xanthopoulos, A., Achillas, C., Michailidis, N., Chatzipanagioti, M., & Kikis, V. (2009). A methodological framework for end-of-life management of electronic products. *Resources, Conservation and Recycling*, *53*(6), 329-339.
130. Idjis, H., Attias, D., Bocquet, J. C., & Richet, S. (2013, September). Designing a sustainable recycling network for batteries from electric vehicles. Development and optimization of scenarios. In *Working Conference on Virtual Enterprises* (pp. 609-618). Springer Berlin Heidelberg.
131. IEEE. (2011). IEEE Recommended Practice for Sizing Lead-Acid Batteries for Stationary Applications. IEEE Power & Energy Society and Stationary Batteries Committee. http://ieeexplore.ieee.org/xpl/mostRecentIssue.jsp?punumber=5751582. Accessed 15 November 2014
132. Infomine. (2015). Manganese Prices and Manganese Price Charts Retrieved from www.infomine.com/investment/metal-prices/manganese/. Accessed 15 July 2015
133. Inside EVs. (2015). Monthly Plug-In Sales Scorecard. http://insideevs.com/monthly-plug-in-sales-scorecard/. Accessed 20 April 2015

134. International Energy Agency. (2011). Technology Roadmap. Electric and plug-in hybrid electric vehicles. Retrieved from http://www.iea.org/publications/freepublications/publication/EV_PHEV_Roadmap.pdf
135. International Institute for Sustainable Development IISD. (2010). Sustainable development and the global copper supply chain: International research team report. Toronto, Canada: Five Winds International.
136. International Organization for Standardization [ISO]. (2006). Environmental management – life cycle assessment – principles and framework (ISO 14044: 2006). Geneva, Switzerland
137. Ishihara, K., Kihira, N., Terada, N., & Iwahori, T. (2002). Environmental burdens of large lithium-ion batteries developed in a Japanese national project. *Central Research Institute of Electric Power Industry, Japan*.
138. Jobin, P., Yamaguchi, J., Balter, G., Ceraso, C.J., Chan, A., Eggers, D., Ellinghorst, A., Flannery, M., McNutley, J.P., Pearson, S. and Pitzer, J.W. (2009). Electric Vehicles. Credit Suisse, Global Equity Research, Energy Technology /Auto Parts & Equipment.
139. Kalhammer, F. R., Kamath, H., Duvall, M., Alexander, M., & Jungers, B. (2009, May). Plug-in hybrid electric vehicles: promise, issues and prospects. In *EVS24 International Battery, Hybrid and Fuel Cell Electric Vehicle Symposium. Stavanger, Norway* (pp. 1-11).
140. Kang, D. H. P., Chen, M., & Ogunseitan, O. A. (2013). Potential environmental and human health impacts of rechargeable lithium batteries in electronic waste. *Environmental science & technology*, *47*(10), 5495-5503.
141. Kang, H. Y., & Schoenung, J. M. (2006). Estimation of future outflows and infrastructure needed to recycle personal computer systems in California. *Journal of hazardous materials*, *137*(2), 1165-1174.
142. Kang, S. H., Lu, W., Gallagher, K. G., Park, S. H., & Pol, V. G. (2011). Study of Li1+ x (Mn4/9Co1/9Ni4/9) 1-xO2 Cathode Materials for Vehicle Battery Applications. *Journal of the Electrochemical Society*, 158(8), A936-A941.
143. Karden, E., Ploumen, S., Fricke, B., Miller, T., & Snyder, K. (2007). Energy storage devices for future hybrid electric vehicles. *Journal of Power Sources*, *168*(1), 2-11.
144. Karlsson, R. (1994, February). LCA as a guide for the improvement of recycling. In *Proceedings of the European Workshop on Allocation in LCA* (pp. 18-28).
145. Kim, S., Hwang, T., & Lee, K. M. (1997). Allocation for cascade recycling system. *The International Journal of Life Cycle Assessment*, *2*(4), 217-222.
146. Kiritsis, D., Bufardi, A., & Xirouchakis, P. (2003, May). Multi-criteria decision aid for product end of life options selection. In *Electronics and the Environment, 2003. IEEE International Symposium on* (pp. 48-53). IEEE
147. Klöpffer, W. (1996). Allocation rule for open-loop recycling in life cycle assessment. *The International Journal of Life Cycle Assessment*, *1*(1), 27-31.
148. Knoepfel, I. (1994, February). Allocation of environmental burdens in life-cycle analysis according to a two-stage procedure. In *Proceedings of the European Workshop on Allocation in LCA, CML, Leiden, Februray* (pp. 24-25).
149. Köhler, A. R., Som, C., Helland, A., & Gottschalk, F. (2008). Studying the potential release of carbon nanotubes throughout the application life cycle. *Journal of Cleaner Production*, *16*(8), 927-937

150. Krieger, E. M., Cannarella, J., & Arnold, C. B. (2013). A comparison of lead-acid and lithium-based battery behavior and capacity fade in off-grid renewable charging applications. *Energy*, *60*, 492-500.
151. Kumar, V., & Sutherland, J. W. (2008). Sustainability of the automotive recycling infrastructure: review of current research and identification of future challenges. *International Journal of Sustainable Manufacturing*, *1*(1), 145-167.
152. Kuo, I. (2011). Tesla to Recycle Electric Car Batteries in Belgium. The New York Times. Retrieved from http://www.batteryrecycling.umicore.com/UBR/media/pressCoverage/2011/show_20110127_NYTimes_BR.pdf. Accessed 21 January 2015
153. Kushnir, D., & Sandén, B. A. (2012). The time dimension and lithium resource constraints for electric vehicles. *Resources Policy*, *37*(1), 93-103.
154. Legal Information Institute. (2015). 49 CFR 173.185 - Lithium cells and batteries. Retrieved from https://www.law.cornell.edu/cfr/text/49/173.185
155. Lemp, J. D., & Kockelman, K. M. (2008). Quantifying the external costs of vehicle use: Evidence from America's top-selling light-duty models. *Transportation research part D: transport and environment*, *13*(8), 491-504.
156. Li, J., Shi, P., Wang, Z., Chen, Y., & Chang, C. C. (2009). A combined recovery process of metals in spent lithium-ion batteries. *Chemosphere*, *77*(8), 1132-1136.
157. Li, L., Dunn, J. B., Zhang, X. X., Gaines, L., Chen, R. J., Wu, F., & Amine, K. (2013). Recovery of metals from spent lithium-ion batteries with organic acids as leaching reagents and environmental assessment. *Journal of Power Sources*, *233*, 180-189.
158. Li, L., Ge, J., Wu, F., Chen, R., Chen, S., Wu, B. (2010). Recovery of cobalt and lithium from spent lithium ion batteries using organic citric acid as leachant. *Journal of Hazardous Materials*, *176* (1-3), 288-293.
159. Linden, D., & Reddy, T. B. (Eds.). (2003). Handbook of batteries. McGraw-Hill
160. London Metal Exchange [LME]. (2012). Pricing and data. Accessed July 2012 from http://www.lme.com/
161. Lowe, M., Tokuoka, S., Trigg, T., & Gereffi, G. (2010). Lithium-ion Batteries for Electric Vehicles: The US Value Chain. *Center on Globalization Governance and Competitiveness, Duke University*.
162. Lu, L., Han, X., Li, J., Hua, J., & Ouyang, M. (2013). A review on the key issues for lithium-ion battery management in electric vehicles. *Journal of power sources*, *226*, 272-288.
163. Madani, A. (2009). HEV, P-HEV& EV Market Trends 2008-2020: Battery is the Key! Presentation by Avicenne Développment for BATTERIES 2009: The International power supply conference and exhibition, Cannes, France. Retrieved from http://www.cars21.com/web/assets/link/Ali%20Madani%20presentation.pdf
164. Majeau-Bettez, G., Hawkins, T. R., & Strømman, A. H. (2011). Life cycle environmental assessment of lithium-ion and nickel metal hydride batteries for plug-in hybrid and battery electric vehicles. *Environmental science & technology*, *45*(10), 4548-4554.
165. Majima, M., Ujiie, S., Yagasaki, E., Koyama, K., & Inazawa, S. (2001). Development of long life lithium ion battery for power storage. *Journal of Power Sources*, *101*(1), 53-59.
166. Mantuano, D. P., Dorella, G., Elias, R. C. A., & Mansur, M. B. (2006). Analysis of a hydrometallurgical route to recover base metals from spent rechargeable batteries by liquid–liquid extraction with Cyanex 272. *Journal of Power Sources*, *159*(2), 1510-1518.

167. Marano, V., & Rizzoni, G. (2008, September). Energy and economic evaluation of PHEVs and their interaction with renewable energy sources and the power grid. Proceedings of the 2008 IEEE International Conference on Vehicular Electronics and Safety. Columbus, OH, USA. September 22-24, 2008, pp. 84-89.
168. Marano, V. , Onori, S. , Guezennec, Y. , Rizzoni, G. , & Madella, N. (2009). Lithium-ion batteries life estimation for plug-in hybrid electric vehicles. 2009 IEEE Vehicle Power and Propulsion Conference, Vols 1-3, 1, 476-483.
169. Matheys, J., & Van Autenboer, W. (2005). SUBAT: sustainable batteries work package 5: overall assessment final public report. *Vrije Universiteit Brussel, Brussels*.
170. Matheys, J., Timmermans, J. M., Van Mierlo, J., Meyer, S., & Van den Bossche, P. (2009). Comparison of the environmental impact of five electric vehicle battery technologies using LCA. *International Journal of sustainable manufacturing*, *1*(3), 318-329.
171. Matheys, J., Van Mierlo, J., Timmermans, J. M., & Van den Bossche, P. (2008). Life-cycle assessment of batteries in the context of the EU Directive on end-of-life vehicles. *International Journal of Vehicle Design*, *46*(2), 189-203.
172. McDowall, J. (2000). Aging factors in battery sizing - Prudent engineering or waste of effort ? Battcon Proceedings, 2000. Retrieved from http://www.battcon.com/PapersScanned2000/McDowallPaper2000.pdf. Accessed 13 November 2014
173. McKinsey & Company. (2014). Moving Toward a Circular Economy. Retrieved from www.mckinsey.com/business-functions/sustainability-and-resource-productivity/our-insights/moving-toward-a-circular-economy. Accessed 4 October 2015.
174. Michaelis, P., & Jackson, T. (2000). Material and energy flow through the UK iron and steel sector: Part 2: 1994–2019. *Resources, Conservation and recycling*, *29*(3), 209-230.
175. Mikolajczak, C., Kahn, M., White, K., & Long, R. T. (2011). Summary of Applicable Codes and Standards. In *Lithium-Ion Batteries Hazard and Use Assessment* (pp. 31-42). Springer US.
176. Minnesota Pollution Control Agency [MN PCA]. (2015). Product stewardship for rechargeable batteries. St. Paul, MN: Minnesota Pollution Control Agency.
177. Mudgal, S., Le Guern, Y., Tinetti, B., Chanoine, A., Pahal, S., & Witte, F. (2011). Comparative Life-Cycle Assessment of nickel-cadmium (NiCd) batteries used in Cordless Power Tools (CPTs) vs. their alternatives nickel-metal hydride (NiMH) and lithium-ion (Li-ion) batteries. Final Report of European Commission. Paris: Bio Intelligence Service.
178. Nagpure, S. C., Downing, R. G., Bhushan, B., Babu, S. S., & Cao, L. (2011). Neutron depth profiling technique for studying aging in Li-ion batteries. *Electrochimica Acta*, *56*(13), 4735-4743.
179. Nan, J., Han, D., & Zuo, X. (2005). Recovery of metal values from spent lithium-ion batteries with chemical deposition and solvent extraction. *Journal of Power Sources*, *152*, 278-284.
180. Narula, C. K., Martinez, R., Onar, O., Starke, M., & Andrews, G. (2011). Final report economic analysis of deploying used batteries in power systems. *Oak Ridge National Laboratory, Oak Ridge, TN*.
181. National Institute for Occupational Safety [NIOSH]. (2007). NIOSH pocket guide to chemical hazards. Retrieved from http://www.cdc.gov/niosh/docs/2005-149/pdfs/2005-149.pdf

182. Nelson, P. A., Bloom, K. G., & I Dees, D. W. (2011). *Modeling the performance and cost of lithium-ion batteries for electric-drive vehicles* (No. ANL-11/32). Argonne National Laboratory (ANL), Argonne, IL (United States).
183. Nelson, P., & Amine, K. (2007). Advanced Lithium-Ion batteries for plug-in hybrid-electric vehicles. Lithium batteries: Research, technology and applications, 203. Retrieved from http://bib.convdocs.org/docs/25/24104/conv_1/file1.pdf#page=216
184. Nemry, F., Leduc, G., & Munoz, A. (2009) Plug-in hybrid and battery-electric vehicles: State of the Research and Development and Comparative Analysis of Energy and Cost Efficiency, Technical Note., Institute for Prospective Technological Studies, Joint Research Centre, European Union. Retrieved from ftp://ftp.jrc.es/pub/EURdoc/JRC54699_TN.pdf
185. Nenadic, N. G., Bussey, H. E., Ardis, P. A., & Thurston, M. G. (2014). Estimation of state-of-charge and capacity of used lithium-ion cells. *International Journal of Prognostics and Health Management*, *5*, 12.
186. Neubauer, J., & Pesaran, A. (2011). The ability of battery second use strategies to impact plug-in electric vehicle prices and serve utility energy storage applications, *Journal of Power Sources*, 196(23), 10351-10358.
187. Neubauer, J., Pesaran, A., Williams, B., Ferry, M., & Eyer, J. (2012). A techno-economic analysis of PEV battery second use: repurposed-battery selling price and commercial and industrial end-user value. SAE Technical Paper (No. 2012-01-0349)
188. New York State Rechargeable Battery Law. (2010). New York Environmental Conservation Law. In: Title 18. Rechargeable Battery Recycling (27-1081 e 27-1811)
189. Newell, S. A., & Field, F. R. (1998). Explicit accounting methods for recycling in LCI. *Resources, Conservation and Recycling*, *22*(1), 31-45.
190. Nicholson, A. L., Olivetti, E. A., Gregory, J. R., Field, F. R., & Kirchain, R. E. (2009, May). End-of-life LCA allocation methods: open loop recycling impacts on robustness of material selection decisions. In *2009 IEEE International Symposium on Sustainable Systems and Technology* (pp. 1-6). IEEE.
191. Nishi, Y. (2001). Lithium-ion secondary batteries; past 10 years and the future. *Journal of Power Sources*, *100*(1), 101-106.
192. Nissan North America, Inc. (2014). http://www.nissanusa.com/electric-cars/leaf/charging-range/battery/. Accessed 19 January 2015
193. Notter, D. A., Gauch, M., Widmer, R., Wager, P., Stamp, A., Zah, R., & Althaus, H. J. (2010). Contribution of Li-ion batteries to the environmental impact of electric vehicles. *Environmental science & technology*, *44*(17), 6550-6556.
194. Oguchi, M., Kameya, T., Yagi, S., & Urano, K. (2008). Product flow analysis of various consumer durables in Japan. *Resources, Conservation and Recycling*, *52*(3), 463-480.
195. Olivetti, E., Gregory, J., & Kirchain, R. (2011). Life Cycle Impacts of Alkaline Batteries With a Focus on End-Of-Life. *Study conducted for the National Electric Manufacturers Association*.
196. Papargyropoulou, E., Lozano, R., Steinberger, J. K., Wright, N., & bin Ujang, Z. (2014). The food waste hierarchy as a framework for the management of food surplus and food waste. *Journal of Cleaner Production*, *76*, 106-115.
197. Park, J., Hong, S. J., Kim, I., Lee, J. Y., & Hur, T. (2011). Dynamic material flow analysis of steel resources in Korea. *Resources, Conservation and Recycling*, *55*(4), 456-462.
198. Parker, C. D. (2001). Lead–acid battery energy-storage systems for electricity supply networks. *Journal of Power Sources*, *100*(1), 18-28.

199. Parks, K., Denholm, P., & Markel, T. (2007). Costs and emissions associated with plug- in hybrid electric vehicle charging in the Xcel Energy Colorado service territory. Technical Report NREL/TP-640-41410, National Renewable Energy Laboratory, Golden, Colorado.
200. Paulino, J. F., Busnardo, N. G., & Afonso, J. C. (2008). Recovery of valuable elements from spent Li-batteries. *Journal of Hazardous Materials*, *150*(3), 843-849.
201. Pauliuk, S., Wang, T., & Müller, D. B. (2011). Moving toward the circular economy: The role of stocks in the Chinese steel cycle. *Environmental science & technology*, *46*(1), 148-154.
202. Pavlov, D., & Petkova, G. (2002). Phenomena That Limit the Capacity of the Positive Lead Acid Battery Plates II. Electrochemical Impedance Spectroscopy and Mechanism of Discharge of the Plate. *Journal of the Electrochemical Society*, *149*(5), A654-A661.
203. Peterson, S. B., Apt, J., & Whitacre, J. F. (2010). Lithium-ion battery cell degradation resulting from realistic vehicle and vehicle-to-grid utilization. *Journal of Power Sources*, *195*(8), 2385-2392.
204. Pistoia, G., Wiaux, J. P., & Wolsky, S. P. (2001). *Used battery collection and recycling* (Vol. 10). Elsevier, London.
205. Plunkert, P. A. (2005). *Aluminum recycling in the United States in 2000*. U.S. Geological Survey.
206. Ramoni, M. O., & Zhang, H. C. (2013). End-of-life (EOL) issues and options for electric vehicle batteries. *Clean Technologies and Environmental Policy*, *15*(6), 881-891.
207. Rantik, M. (1999). Life cycle assessment of five batteries for electric vehicles under different charging regimes. KFB-Meddelande, vol 28. Chalmers Univ. of Technology, Göteborg, Sweden
208. Reichmuth, D. S., Lutz, A. E., Manley, D. K., & Keller, J. O. (2012). Comparison of the technical potential for hydrogen, battery electric, and conventional light-duty vehicles to reduce greenhouse gas emissions and petroleum consumption in the United States. *International Journal of Hydrogen Energy*, *38*(2), 1200-1208.
209. Richa, K., Babbitt, C. W., Gaustad, G., & Wang, X. (2014). A future perspective on lithium-ion battery waste flows from electric vehicles. *Resources, Conservation and Recycling*, *83*, 63-76.
210. Richa, K., Babbitt, C. W., Nenadic, N. G., & Gaustad, G. (2015). Environmental trade-offs across cascading lithium-ion battery life cycles. *The International Journal of Life Cycle Assessment*, *20*, 1-16.
211. Richter, B., Goldston, D., Crabtree, G., Glicksman, L., Goldstein, D., Greene, D., Kammen, D., Levine, M., Lubell, M., Savitz, M., & Sperling, D. (2008). Energy future: Think efficiency. *American Physical Society: College Park, MD*. Retrieved from http://www.aps.org/energyefficiencyreport/report/aps-energyreport.pdf
212. Ritchie, A., & Howard, W. (2006). Recent developments and likely advances in lithium-ion batteries. *Journal of Power Sources*, *162*(2), 809-812.
213. Robinson, B. H. (2009). E-waste: an assessment of global production and environmental impacts. *Science of the total environment*, *408*(2), 183-191.
214. Rosenbaum, R.K., Bachmann, T.M., Gold, L.S., Huijbregts, M.A., Jolliet, O., Juraske, R., Koehler, A., Larsen, H.F., MacLeod, M., Margni, M. and McKone, T.E. (2008). USEtox—the UNEP-SETAC toxicity model: recommended characterisation factors for human toxicity and freshwater ecotoxicity in life cycle impact assessment. *The International Journal of Life Cycle Assessment*, *13*(7), 532-546.

215. Rossi, V., Cleeve-Edwards, N., Lundquist, L., Schenker, U., Dubois, C., Humbert, S., & Jolliet, O. (2015). Life cycle assessment of end-of-life options for two biodegradable packaging materials: sound application of the European waste hierarchy. *Journal of Cleaner Production*, *86*, 132-145.
216. Ruhrberg, M. (2006). Assessing the recycling efficiency of copper from end-of-life products in Western Europe. *Resources, Conservation and Recycling*, *48*(2), 141-165.
217. Rydberg, T. (1995). Cascade accounting in life cycle assessment applied to polymer recycling. *Polymer Recycling(UK)*, *1*(4), 233-241.
218. Rydh, C. J. (1999). Environmental assessment of vanadium redox and lead-acid batteries for stationary energy storage. *Journal of power sources*, *80*(1), 21-29.
219. Rydh, C. J., & Sandén, B. A. (2005). Energy analysis of batteries in photovoltaic systems. Part I: Performance and energy requirements. *Energy conversion and management*, *46*(11), 1957-1979.
220. Rydh, C. J., & Karlström, M. (2002). Life cycle inventory of recycling portable nickel–cadmium batteries. *Resources, Conservation and Recycling*, *34*(4), 289-309.
221. Saevarsdottir, G., Tao, P. C., Stefansson, H., & Harvey, W. (2014). Potential use of geothermal energy sources for the production of lithium-ion batteries. *Renewable Energy*, *61*, 17-22.
222. Samaras, C., & Meisterling, K. (2008). Life cycle assessment of greenhouse gas emissions from plug-in hybrid vehicles: implications for policy. *Environmental science & technology*, *42*(9), 3170-3176.
223. Sathre, R., Scown, C. D., Kavvada, O., & Hendrickson, T. P. (2015). Energy and climate effects of second-life use of electric vehicle batteries in California through 2050. *Journal of Power Sources*, *288*, 82-91.
224. Saxena, S., Le Floch, C., MacDonald, J., & Moura, S. (2015). Quantifying EV battery end-of-life through analysis of travel needs with vehicle powertrain models. *Journal of Power Sources*, *282*, 265-276.
225. Shanghai Metals Market. (2012). Minor metals prices. Accessed July 2012 from http://www.metal.com/metals/minor-metals/prices
226. Shiau, C. S. N., Samaras, C., Hauffe, R., & Michalek, J. J. (2009). Impact of battery weight and charging patterns on the economic and environmental benefits of plug-in hybrid vehicles. *Energy Policy*, *37*(7), 2653-2663.
227. Shin, S. M., Kim, N. H., Sohn, J. S., Yang, D. H., & Kim, Y. H. (2005). Development of a metal recovery process from Li-ion battery wastes. *Hydrometallurgy*, *79*(3), 172-181.
228. Shrestha, S., Martin, S., & Zhang, J. (2010). Industry Note. Alternative Energy and Infrastructure. Lazard Capital Markets (March, 2010). Retrieved from http://www.eosenergystorage.com/articles/LazardMarketReport%202010-03.pdf
229. Sibley, S. F. (2011). Overview of Flow Studies for Recycling Metal Commodities in the United States. U.S. Geological Survey. Retrieved from http://pubs.usgs.gov/circ/circ1196-AA/
230. Slack, R. J., Gronow, J. R., & Voulvoulis, N. (2005). Household hazardous waste in municipal landfills: contaminants in leachate. *Science of the total environment*, *337*(1), 119-137.
231. Soloveichik, G. L. (2011). Battery technologies for large-scale stationary energy storage. *Annual review of chemical and biomolecular engineering*, *2*, 503-527.

232. Spotnitz, R. (2003). Simulation of capacity fade in lithium-ion batteries. *Journal of Power Sources*, *113*(1), 72-80.
233. Srinivasan, V. (2008). Batteries for Vehicular Applications. *AIP conference proceedings, 1044p*. 283. Retrieved from http://bestar.lbl.gov/venkat/files/batteries-for-vehicles.pdf
234. Srinivasan, V., & Lipp, L. (2003). Report on the Electrolytic Industries for the Year 2002. *Journal of the Electrochemical Society*, *150*(12), K15-K38.
235. Standridge, C. & Corneal, L. (2014).Remanufacturing, Repurposing, and Recycling of Post-Vehicle-Application Lithium-Ion Batteries. Report 12-20. San Jose, CA: Mineta National Transit Research Consortium, June 2014. Retrieved from http://transweb.sjsu.edu/PDFs/research/1137-post-vehicle-Li-Ion-recycling.pdf
236. Steinmetz, J., & Shankar, R. (March, 2008). Autos & Auto Related: Plug in Hybrids: The Next Automotive Revolution. New York: Morgan Stanley Research, North America, p. 13.
237. Steubing, B., Böni, H., Schluep, M., Silva, U., & Ludwig, C. (2010). Assessing computer waste generation in Chile using material flow analysis. *Waste management*, *30*(3), 473-482.
238. Streicher-Porte, M., Marthaler, C., Böni, H., Schluep, M., Camacho, A., & Hilty, L. M. (2009). One laptop per child, local refurbishment or overseas donations? Sustainability assessment of computer supply scenarios for schools in Colombia. *Journal of Environmental Management*, *90*(11), 3498-3511.
239. Streicher-Porte, M., Widmer, R., Jain, A., Bader, H. P., Scheidegger, R., & Kytzia, S. (2005). Key drivers of the e-waste recycling system: Assessing and modelling e-waste processing in the informal sector in Delhi. *Environmental Impact Assessment Review*, 25(5), 472-491.
240. Sullivan, J. L., & Gaines, L. (2012). Status of life cycle inventories for batteries. *Energy Conversion and Management*, *58*, 134-148.
241. Sullivan, J. L., Gaines, L., & Burnham, A. (2011). Role of recycling in the life cycle of batteries. In *TMS 2011 140th annual meeting and exhibition, materials processing and energy materials*.
242. Sumitomo Corp. (2011). Aiming to initiate second-life business for electric car batteries. http://www.sumitomocorp.co.jp/english/business/article/id=263. Accessed 25 January 2015
243. Sustainable Energy Ireland [SEI]. (2007). Hybrid electric and battery electric vehicles-technology, costs and benefits. ILTP Consulting. Retrieved from http://www.seai.ie/News_Events/Press_Releases/Costs_and_benefits.pdf. Accessed 5 April 2014
244. Swain, B., Jeong, J., Lee, J. C., Lee, G. H., & Sohn, J. S. (2007). Hydrometallurgical process for recovery of cobalt from waste cathodic active material generated during manufacturing of lithium ion batteries. *Journal of Power Sources*, *167*(2), 536-544.
245. Tanaka, T., Ohta, K., & Arai, N. (2001). Year 2000 R&D status of large-scale lithium-ion secondary batteries in the national project of Japan. *Journal of power sources*, *97*, 2-6.
246. U.S Environmental Protection Agency and U.S. Department of Energy. (2013). www.fueleconomy.gov- The official US government source for fuel economy information. Retrieved from http://www.fueleconomy.gov/
247. U.S. Department of Energy. (2010). Final Environmental Assessment for Compact Power, Inc. Electric Drive Vehicle Battery and Component Manufacturing Initiative Application, Holland, Michigan. U.S. Department of Energy, National Energy Technology Laboratory. Retrieved from http://energy.gov/sites/prod/files/nepapub/nepa_documents/RedDont/EA-1709-FEA-2010.pdf. Accessed 20 October 2014

248. U.S. DOE. (July, 2010). The recovery Act: Transforming America's transportation sector. Batteries and electric vehicles. Retrieved from http://www.whitehouse.gov/files/documents/Battery-and-Electric-Vehicle-Report-FINAL.pdf
249. U.S. EPA. (2012). Lithium-Ion Batteries and Nanotechnology for Electric Vehicles: A Life Cycle Assessment. EPA 744-R-12-001. U.S. EPA, Washington DC
250. US Department of Transportation (USDOT). (2014). U.S. Department of Transportation Issues New Standards to Improve Safety of Lithium Battery Transportation. Retrieved from https://www.transportation.gov/briefing-room/us-department-transportation-issues-new-standards-improve-safety-lithium-battery
251. US Department of Transportation (USDOT). (December, 2015). DOT-Special Permit 16474, Pipeline and Hazardous Materials Safety Administration. Retrieved from http://www.phmsa.dot.gov/staticfiles/PHMSA/SPA_App/OfferDocuments/SP16474_201505 0361.pdf
252. US EPA. (2009). National Primary Drinking Water Regulations. Retrieved from http://water.epa.gov/drink/contaminants/upload/mcl-2.pdf
253. US EPA. (2015a). Advancing Sustainable Materials Management: Facts and Figures. Retrieved from www.epa.gov/solidwaste/nonhaz/municipal/pubs/2013_advncng_smm_rpt.pdf. Accessed December 2015.
254. US EPA. (2015b). Sustainable Materials Management: Non-Hazardous Materials and Waste Management Hierarchy. Retrieved from www.epa.gov/smm/sustainable-materials-management-non-hazardous-materials-and-waste-management-hierarchy. Accessed January 2016.
255. US EPA. (2016). Food Recovery Hierarchy. Retrieved from www.epa.gov/sustainable-management-food/food-recovery-hierarchy. Accessed January 2016.
256. US GPO. (1996). Public Law 104–142-Mercury Containing and Rechargeable Battery Management Act. Retrieved from http://www.gpo.gov/fdsys/pkg/PLAW-104publ142/content-detail.html
257. USABC. (2014). 2014 Recommended Practice for Recycling of xEV Electrochemical Energy Storage Systems. United States Advanced Battery Consortium Report. Retrieved from http://uscar.org/guest/teams/68/USABC-Battery-Recycling-Group
258. USGS. (2009). Lithium, U.S. Geological Survey, Mineral Commodity Summaries 2009. Retrieved from http://minerals.usgs.gov/minerals/pubs/commodity/lithium/mcs-2009-lithi.pdf
259. USGS. (2012). Mineral Commodity Summaries 2012. U.S. Geological Survey 2012.
260. USGS. (2015). Mineral Commodity Summaries 2015. U.S. Geological Survey 2015.
261. Van den Bossche, P., Vergels, F., Van Mierlo, J., Matheys, J., & Van Autenboer, W. (2006). SUBAT: An assessment of sustainable battery technology. *Journal of power sources*, 162(2), 913-919.
262. Van Haaren, R. (2011). Assessment of Electric Cars' Range Requirements and Usage Patterns based on Driving Behavior recorded in the National Household Travel Survey of 2009. Earth and Environmental Engineering Department, Columbia University, Fu Foundation School of Engineering and Applied Science, New York
263. Vimmerstedt, L. J., Ring, S., & Hammel, C. J. (1995). Current status of environmental, health, and safety issues of lithium-ion electric vehicle batteries (No. NREL/TP--463-7673).

National Renewable Energy Lab., Golden, CO (United States). Retrieved from http://www.osti.gov/energycitations/product.biblio.jsp?osti_id=135056
264. Viswanathan, V. V., & Kintner-Meyer, M. (2011). Second use of transportation batteries: Maximizing the value of batteries for transportation and grid services. *IEEE Transactions on vehicular technology*, *60*(7), 2963-2970.
265. Walls, M. (2006). Extended producer responsibility and product design: economic theory and selected case studies. Paris: OECD.
266. Wang, X., Gaustad, G., Babbitt, C. W., Bailey, C., Ganter, M. J., & Landi, B. J. (2014a). Economic and environmental characterization of an evolving Li-ion battery waste stream. *Journal of environmental management*, *135*, 126-134.
267. Wang, X., & Gaustad, G. (2012). Prioritizing material recovery for end-of-life printed circuit boards. *Waste Management*, *32*(10), 1903-1913.
268. Wang, X., Gaustad, G., & Babbitt, C. W. (2015). Targeting high value metals in lithium-ion battery recycling via shredding and size-based separation. *Waste Management*.
269. Wang, X., Gaustad, G., Babbitt, C. W., & Richa, K. (2014b). Economies of scale for future lithium-ion battery recycling infrastructure. *Resources, Conservation and Recycling*, *83*, 53-62.
270. Watabe, T., & Mori, M. (2011). LIB materials industry. Automotive LIB materials get set for growth phase in 2011. Global Markets Research. Deutsche Bank Group. Retrieved from http://www.fullermoney.com/content/2012-02-02/LiB12611.pdf
271. Webster, H. (2010). Fire protection for shipment of lithium batteries in aircraft cargo compartments. U.S. Department of Transportation.
272. Wellbeloved, D. B., Craven, P. M., & Waudby, J. W. (1990). Manganese and manganese alloys. *Ullmann's Encyclopedia of Industrial Chemistry*.
273. Werner, F., Althaus, H. J., Richter, K., & Scholz, R. W. (2007). Post-consumer waste wood in attributive product LCA. *The International Journal of Life Cycle Assessment*, *12*(3), 160-172.
274. Widmer, R., Oswald-Krapf, H., Sinha-Khetriwal, D., Schnellmann, M., & Böni, H. (2005). Global perspectives on e-waste. *Environmental Impact Assessment Review*, *25*(5), 436-458.
275. Williams, B. (2011). Plug-In-Vehicle Battery Second Life: the Effect of Post-Vehicle, Distributed-Grid Energy Storage Value on Battery-Lease Payments. Transportation Sustainability Research Center, University of California, Berkeley, CA
276. Williams, B. D., & Lipman, T. E. (2010). Strategy for Overcoming Cost Hurdles of Plug-In-Hybrid Battery in California. *Transportation Research Record: Journal of the Transportation Research Board*, *2191*(1), 59-66.
277. Williams, E., Kahhat, R., Allenby, B., Kavazanjian, E., Kim, J., & Xu, M. (2008). Environmental, social, and economic implications of global reuse and recycling of personal computers. *Environmental Science & Technology*, *42*(17), 6446-6454.
278. World Health Organization [WHO]. (2008). Guidelines for drinking-water quality - Volume 1: Recommendations. Retrieved from www.who.int/water_sanitation_health/dwq/gdwq3rev/en/. Accessed 15 January 2016
279. Xu, T., Wang, W., Gordin, M. L., Wang, D., & Choi, D. (2010). Lithium-ion batteries for stationary energy storage. The Journal of the Minerals, Metals & Materials Society, *62*(9), 24-30.

280. Xu, J., Thomas, H. R., Francis, R. W., Lum, K. R., Wang, J., & Liang, B. (2008). A review of processes and technologies for the recycling of lithium-ion secondary batteries. *Journal of Power Sources*, *177*(2), 512-527.
281. Yaksic, A., & Tilton, J. E. (2009). Using the cumulative availability curve to assess the threat of mineral depletion: The case of lithium. *Resources Policy*, 34(4), 185-194.
282. Yang, Z., Mei, Z., Xu, F., Yao, Y., Zhang, W., Qi, W., & Zhao, T. (2013). Different types of MnO2 recovered from spent LiMn2O4 batteries and their application in electrochemical capacitors. *Journal of Materials Science*, *48*(6), 2512-2519.
283. Yoshida, A., Tasaki, T., & Terazono, A. (2009). Material flow analysis of used personal computers in Japan. *Waste management*, 29(5), 1602-1614.
284. Yoshio, M., & Noguchi, H. (2009). A review of positive electrode materials for lithium-ion batteries. In *Lithium-Ion Batteries* (pp. 9-48). Springer New York.
285. Zackrisson, M., Avellán, L., & Orlenius, J. (2010). Life cycle assessment of lithium-ion batteries for plug-in hybrid electric vehicles–Critical issues.*Journal of Cleaner Production*, *18*(15), 1519-1529.
286. Zhang, Y., Wang, C. Y., & Tang, X. (2011). Cycling degradation of an automotive LiFePO4 lithium-ion battery. *Journal of Power Sources*, *196*(3), 1513-1520.
287. Zou, H., Gratz, E., Apelian, D., & Wang, Y. (2013). A novel method to recycle mixed cathode materials for lithium-ion batteries. *Green Chemistry*, *15*(5), 1183-1191.